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#### A state-of-the-art review

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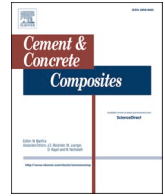
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# Thixotropic structural build-up of cement-based materials: A state-of-the-art review

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## ABSTRACT

Thixotropic structural build-up is an intrinsic property of fresh cementitious materials, playing a significant role in pumpability, stability and formwork filling of concrete, as well as buildability in 3D printing. This paper presents a critical review on the thixotropic structural build-up of cement-based materials from the viewpoints of origins, evaluation methods, influencing factors and applications. Recent research indicates that the origin of structural build-up of cementitious materials is a combining result of colloidal interactions and chemical hydration, with three stages of colloidal network percolation, rigid percolation and rigidification. The evolution of static yield stress and storage modulus can be used to evaluate the structural build-up of fresh cementitious materials, each describing different aspects. Increasing particle volume fraction, decreasing surface coverage and reducing maximum packing fraction exhibit a positive effect on the increase in the structural build-up. Based on the relationships with performance parameters, the thixotropic structural build-up can be used to predict formwork casting, numerical simulations and 3D concrete printing. Furthermore, on-demand stiffening can be obtained by magneto-controlled structural build-up of cementitious materials containing magnetizable particles.

## 1. Introduction

Fresh cementitious materials can be regarded as concentrated colloidal suspensions with solid particles dispersed in water solution. The rheological properties of fresh cementitious suspensions immediately after mixing can be described by two fundamental physical parameters, i.e., (dynamic) yield stress and plastic viscosity [1–3]. The yield stress and plastic viscosity are of great importance in guiding mixture proportion design [4,5], describing flowability and workability [6,7], evaluating pumpability [8,9], and predicting formwork casting of cement-based materials [10–12]. Due to the colloidal interactions between solid particles and chemical hydration of cementitious materials, the internal microstructure of the suspension gradually evolves over time. This intrinsic property results in cementitious suspensions generally exhibiting time-dependent thixotropic behavior. With the application of self-compacting concrete (SCC) and the development of digital fabrication with concrete, the thixotropic behavior of cementitious materials has attracted widespread research attention [13–17].

The concept of thixotropy was first introduced by Freundlich [18],

described by a continuous decrease of viscosity with time when applying a shear stress to a suspension at rest and then subsequent recovery of viscosity over time when the shear stress is removed [14]. This means that thixotropy is characterized by two aspects, i.e., structural build-up of the suspension at rest and structural breakdown due to applied shearing [19,20]. Thixotropy is a reversible process. In the case of cementitious materials elapsed with time, reversible change due to colloidal flocculation and partial irreversible evolution of internal microstructure owing to chemical hydration exist simultaneously [21, 22]. Therefore, structural build-up involving both reversible and irreversible processes, is a more precise term than thixotropy for cement-based materials at rest state [23]. Note that chemical hydration induced structural build-up and breakdown, although shearing irreversibly destroys the bridges between particles microscopically, can be perceived macroscopically as a partial reversible process due to the reformation of new bridges as a result of the early hydration reaction. The early structural build-up has a significant influence on flowability and pumpability [24–27], and plays a leading role in stability and formwork filling of concrete during casting [28–30].

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Due to its high potential in applications, the amount of research and knowledge of thixotropic structural build-up of cement-based materials has increased significantly in recent years. In the present paper, the origins and evaluation methods of structural build-up of cementitious materials are illustrated. The influences of mix proportion parameters (such as water-to-binder ratio, cementitious materials, aggregates and chemical admixtures) and external factors such as temperature and magnetic field on the structural build-up of cementitious materials are discussed in detail. The applications of structural build-up in guiding formwork casting, numerical simulation, 3D printing and active stiffening control are also reviewed.

## 2. Origins

Fresh cementitious material is a suspension undergoing physical and chemical changes. At rest, several forces exist in the system, such as Brownian motion, van der Waals attractive forces, electrostatic double layer forces or steric hindrance forces, gravitational and inertial forces [16,31]. Brownian motion is a kind of thermal randomizing force. It is only noticeable for particles with size smaller than  $1\ \mu\text{m}$ . In this case, the influence of Brownian force is comparable to that of gravity. In contrast, the thermal activation on larger particles is negligible. Van der Waals attractive forces, containing Keesom, Debye and London interactions, originate from the moment of dipoles of atoms or molecules [32]. In the case of electrostatic repulsive force, it is generated by the electrical double layer of particles. At long intermolecular distances, the van der Waals attraction dominates while at short distances the steep Born repulsion originating from the overlapping electron clouds of molecules dominates [33]. For cementitious paste with small values of zeta potential (typically ranging from  $-15$  to  $20\ \text{mV}$  [34,35]), the repulsive electrostatic double layer interaction is not strong enough to overcome the van der Waals attractive forces [31,36].

The increase in yield stress of cement suspensions is generally regarded as the effect of colloidal interactions between solid particles and the nucleation of early hydration products at their contact points [16,37]. At the end of mixing or shearing, cement particles are highly dispersed in the suspension. Due to the hydrogen/ionic bonding and colloidal interactions, cementitious particles start to agglomerate, forming a soft colloidal structure [15,19]. It should be noted that this weak network has an ability to resist external shear stress, displaying an elastic response and thereby a shear modulus. Meanwhile, early hydration products such as metastable ettringite and C-S-H are formed as bridges around cement particles [16,38–40], resulting in a stronger connection between the particles. When applying an additional shearing, the interparticle bridges of the early hydration products can be broken down and the particles separate again. In this case, the surfaces of un-hydrated cement particles are exposed to a water environment, with ions dissolving and new hydration products forming again [41,42]. When the shearing state is stopped, however, cement particles flocculate again [33]. With increasing resting time, the agglomeration structure accumulates under the influence of van der Waals attraction forces and forms a network structure. Simultaneously, the nucleation of early hydration products strengthens the connections between cement particles within the network, resulting in the transformation from colloidal interactions to more rigid connections [16,43]. The increase in the bridges of hydration products due to the progress of chemical hydration results in further improvement of structural strength, which can be explained by the combination effect of the increase in double layer compression and surface roughness, the consumption of superplasticizer molecules, as well as the steric particle bridging [44,45].

Overall, the structural build-up of cementitious materials is the combined result of physical interactions and chemical hydration. The physicochemical process can be summarized in the following three stages, i.e., formation of network due to colloidal interactions (Colloidal network percolation), formation of early hydration products such as C-S-H and ettringite bridges between cement particles (Rigid

percolations), and the strengthening of the inter-particle connections (Rigidification). This mechanism can be clearly observed from the schematic diagram of thixotropic structural build-up of cementitious materials containing superplasticizer in Fig. 1.

## 3. Evaluation methods

The widely used approaches to evaluate the structural build-up of fresh cementitious materials include static yield stress test, creep recovery test, and small amplitude oscillatory shear (SAOS) test. Generally, the static yield stress and creep recovery tests are considered destructive measurements, whereas the SAOS test is a non-destructive method. In this section, the fundamental principles of these methods are illustrated, and some conventional workability test methods are also briefly demonstrated.

### 3.1. Static yield stress

#### 3.1.1. Measurements

Static yield stress is defined as the minimum shear stress required for initiating flow. Essentially, it is associated with the links and connections between agglomerate structures and particles [16]. Therefore, the evolution of static yield stress can be used to evaluate the intensity of structural build-up of fresh cement-based materials.

There are several testing modes to obtain the static yield stress, such as shear rate-controlled, shear strain-controlled and shear stress-controlled [16,23,33,38,46]. The most popular mode is the shear rate-controlled method, i.e., constant shear rate method or stress growth test. In the context of cementitious materials, the commonly used shear rate ranges from  $0.001\ \text{s}^{-1}$  to  $0.05\ \text{s}^{-1}$  for cement paste [37,47–49], while in the case of mortar and concrete, a relatively high shear rate ( $0.01$ – $0.1\ \text{s}^{-1}$ ) is usually adopted [38,50,51]. The constant shear rate test results mostly depend on the geometry of rheometer, mixture proportions, shear rate and duration of pre-shearing and rheological test. Indeed, Yuan et al. [23] evaluated the effects of shear rates ( $0.001$ ,  $0.003$ ,  $0.005$ ,  $0.008$ ,  $0.01$  and  $0.02\ \text{s}^{-1}$ ) on the measured static yield stress of cement paste. A representative result for cement paste with water-to-cement (w/c) ratio of  $0.45$  and without superplasticizer is shown in Fig. 2. It can be seen that no peak value was reached at shear rate of  $0.001\ \text{s}^{-1}$  during the entire test duration, but the ultimate shear stress, although time-dependent, was much higher than that obtained under other shear rates. This is possibly due to the continuous formation of internal structure with elapsed time, which dominates the structural breakdown induced by the low shear rate. In this case, the determination of the static yield stress will be more subjective. Lower static yield stress and less time to reach the peak (plateau) were recognized for higher shear rate, indicating that more structure of cement paste might be disturbed. It is recommended that a shear rate of  $0.005\ \text{s}^{-1}$  can be used in stress growth test to monitor the structural build-up of fresh cement paste [23].

As aforementioned, the static yield stress cannot be successfully obtained when selecting an inappropriate shear rate. This phenomenon particularly occurs in the constant shear rate test for stiff cementitious materials such as 3D printing materials. In this case, some other approaches are proposed to calculate the static yield stress in the constant shear rate test. For example, Ivanova and Mechtcherine [48] stated that the static yield stress can be determined by the shear stress at the end of linear increase in shear rate-shear strain curve, instead of the maximum shear stress on flow onset. Given that the effective shear rate needs some time to reach the applied shear rate for very stiff cementitious materials, Nerella et al. [52] suggested that the static yield stress test can be conducted by inputting a constant effective strain  $\gamma_{ef}$ , which is higher than the minimum strain to ensure flow-onset ( $\gamma_{ef} \geq \gamma_{ef,min} \approx 1.5$  units for the pastes in their study). This approach is based on the hypothesis that the variation of shear rate can be compensated by the test duration, as long as the applied shear strain is constant. This transforms the constant shear

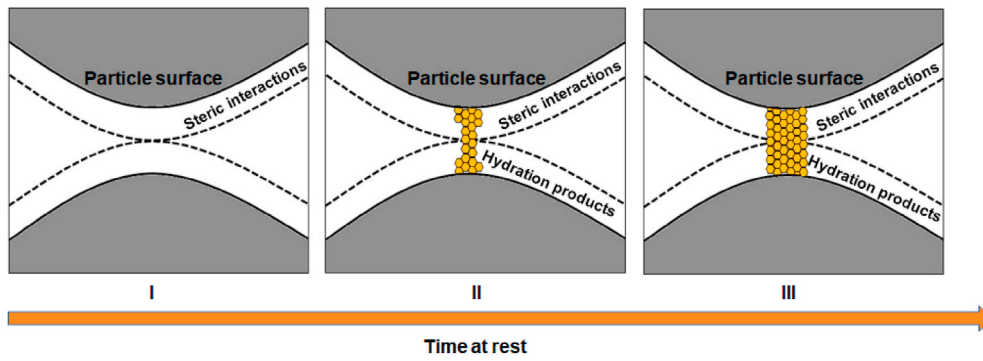


Fig. 1. Schematic diagram of structural build-up of cementitious suspensions with superplasticizer (adapted from Ref. [38]).

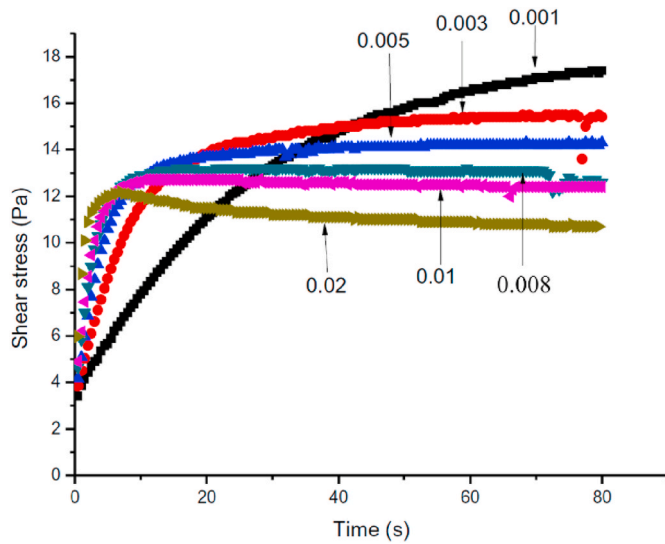


Fig. 2. Static yield stress test of cement paste at different shear rates [23].

rate test to a strain-based analysis approach. According to this method, the static yield stress can be determined by the maximum shear stress obtained. This strain-based analysis approach can be used to capture the structural build-up of very stiff materials.

### 3.1.2. Structural build-up vs evolution of static yield stress

The structural build-up rate can be evaluated by the growth rate of the static yield stress. Assuming that the Bingham model is sufficient to describe the rheological behavior of cementitious materials at steady flow state, according to Roussel [19], the reversible character of thixotropy can be expressed by a kinetic evolution of an internal structural parameter  $\lambda$  in Eq. (1).

$$\begin{cases} \tau_0(t) = (1 + \lambda)\tau_0 + \mu_p \dot{\gamma} \\ \frac{\partial \lambda}{\partial t} = \frac{1}{T} - \alpha \lambda \dot{\gamma} \end{cases} \quad (1)$$

where  $\tau_0(t)$  and  $\tau_0$  are the static yield stress at resting time  $t$  and initial static yield stress, respectively,  $\mu_p$  is the plastic viscosity,  $\dot{\gamma}$  is the shear rate,  $T$  is the flocculation characteristic time, and  $\alpha$  is the de-flocculation rate under shear, with the value equal to 0.006 for cement-based suspensions [19].  $\lambda$  describes the flocculation state of the material, which is regarded as zero immediately after mixing and increases linearly with resting time. The reversible character of the internal structure is expressed by the same build-up through a characteristic flocculation time, while breakdown under shear through a de-flocculation rate. Assuming that the flocculation time is much longer than the

characteristic time of de-flocculation, and the static yield stress increases linearly during the dormant period, the evolution of static yield stress with time can be expressed by Eq. (2).

$$\tau_0(t) = \tau_0 + A_{thix} \cdot t \quad (2)$$

with

$$A_{thix} = \frac{\tau_0}{T} \quad (3)$$

where  $A_{thix}$  is defined as flocculation (structuration) rate, i.e., thixotropic structural build-up rate. This parameter can successfully express the experimental observations. A typical classification of SCC and its corresponding flocculation rate is depicted in Table 1.

After the linear increase period lasting up to 60 min, however, the structural build-up speeds up due to cement hydration. In this case, Perrot et al. [47] proposed an exponential static yield stress evolution model describing a smooth transition from the initial linear increase to an exponential evolution, as expressed by Eq. (4).

$$\tau_0(t) = \tau_0 + A_{thix} \cdot t_c (e^{t/t_c} - 1) \quad (4)$$

where  $t_c$  is a characteristic time to obtain the best fit with experimental values. This model asymptotically tends to Roussel's model prior to  $t_c$ . By considering the changes of solid volume fraction and particle packing due to cement hydration, Lecompte and Perrot [37] provided a theoretical framework for this non-linear model, which can be expressed as:

$$\tau_0 = m \frac{A_0 a^* \phi^2 (1 + \chi \alpha(t))^{4/3} (\phi(1 + \chi \alpha(t)) - \phi_{perc})}{d_0^3 H^2 \phi_m (\phi_m - \phi(1 + \chi \alpha(t)))} + A_{thix} \cdot t \quad (5)$$

where  $m$  is a pre-factor, depending on the particle size distribution,  $d$  is the particle average diameter,  $a^*$  is the radius of curvature of the contact points,  $H$  is the surface to surface separation distance at contact points,  $A_0$  is the Hamaker constant,  $\phi_{perc}$  is the percolation volume fraction,  $\phi$  is the solid volume fraction,  $\phi_m$  is the maximum volume fraction of particles,  $\alpha(t)$  is an indicator of the hydration degree, and  $\chi$  is a parameter indicating hydrated cement volume [37]. This model extends the linear model to predict the evolution of yield stress over longer time spans, and the effects of both resting time and chemical hydration are taken into account.

In addition, Kruger et al. [53] proposed a bi-linear thixotropy model to correlate the evolution of static yield shear stress and the structural build-up by considering re-flocculation rate ( $R_{thix}$ ) and structuration rate

Table 1  
Classification of SCC and its corresponding flocculation rate [19].

SCC type	Flocculation rate $A_{thix}$ (Pa/s)
Non-thixotropic SCC	Less than 0.1
Thixotropic SCC	Between 0.1 and 0.5
Highly thixotropic SCC	Higher than 0.5

( $A_{thix}$ ), as depicted in Fig. 3. The mathematical expression of the bi-linear model can be described as:

$$\begin{cases} \tau_S(t) = \tau_{D,i} + R_{thix} \cdot t, & t \leq t_{rf} \\ t_{rf} = \frac{\tau_{S,i} - \tau_{D,i}}{R_{thix}} \\ \tau_S(t) = \tau_{S,i} + A_{thix} \cdot t, & t > t_{rf} \end{cases} \quad (6)$$

where  $\tau_S(t)$  is the static yield shear stress of the concrete at time  $t$  after agitation,  $\tau_{D,i}$  and  $\tau_{S,i}$  are the initial dynamic and static yield stress of the concrete, respectively, and  $t_{rf}$  is the time period of re-flocculation. This model distinguishes the physical and chemical influences on the microstructure of a material, denoted by re-flocculation and structuration, respectively, and can be used to describe the thixotropic behavior of 3D printing concrete [53–55].

### 3.2. Creep recovery test

In the case of non-applicable of stress growth test in evaluating highly thixotropic materials, Qian and Kawashima [56,57] used a creep recovery test to measure the static yield stress of cement paste. The principle of this method is based on the fact that the response of a material to a removal of an applied constant shear stress for a period is material-dependent. For viscoelastic material such as fresh cement paste, if the applied shear stress is sufficiently low, the fresh cement paste exhibits viscoelastic solid-like behavior, resulting in an increase in viscosity. When the applied stress is higher than a critical stress, however, the fresh cement paste shows viscoelastic liquid-like behavior and starts to flow, exhibiting a decrease in viscosity. This critical strain where viscosity bifurcates is determined as the static yield stress. Qian and Kawashima [56] stated that the static yield stress obtained from the creep recovery test showed a good agreement with the critical strain determined by oscillatory amplitude strain sweep test. Nevertheless, the creep recovery test is not a convenient method to monitor the evolution of structural build-up of cementitious materials over time.

### 3.3. SAOS test

The small amplitude oscillatory shear (SAOS) test to monitor the structural build-up is generally referred as the time-sweep test. By applying a continuous sinusoidal shear strain within the linear viscoelastic region (LVER), the responses of the material such as storage modulus ( $G'$ ), loss modulus ( $G''$ ) and phase angle ( $\delta$ ) can be obtained,

and these parameters follow the relation:

$$\tan \delta = \frac{G''}{G'} \quad (7)$$

where  $G'$  indicates the solid-like (or elastic) property and  $G''$  represents the liquid-like (or viscous) behavior. Higher storage modulus and lower phase angle indicate higher stiffness of the material. The upper limit of LVER for cementitious paste is generally believed to be in the order of  $10^{-4}$  [58], and thus during the time sweep test, the shear strain is generally within the range of  $10^{-6}$  to  $10^{-4}$  at constant frequency (e.g., 1 Hz). This small shear strain ensures the integrity of internal microstructure, and thus the SAOS test can be considered as a non-destructive method.

Based on the principle of the SAOS test, Mostafa and Yahia [59] defined two parameters, i.e., percolation time and rigidification rate, to quantitatively characterize the structural build-up of cementitious paste. The percolation time reflects the formation of internal microstructure due to the colloidal interactions, and the rigidification rate indicates the structural evolution because of the chemical hydration. These two parameters provide reliable indicators to describe the structural build-up of cementitious paste considering the physical and chemical points. Furthermore, the authors [21] proposed a semi-empirical model to correlate these parameters with the microstructural characteristics. The model of the percolation time ( $t_{perc}$ ) is described as:

$$\begin{cases} t_{perc} = Ae^{B\alpha_r} \\ \alpha_r = \frac{S_d^{n_1}}{I_C^{m_2} \cdot f_C^{m_3}} \end{cases} \quad (8)$$

The model of the rigidification rate ( $R_{rigid}$ ) is expressed as:

$$\begin{cases} R_{rigid} = A^* e^{B^* \alpha_G} \\ \alpha_G = \frac{I_C^{m_1} \cdot K_B^{m_1} \cdot \phi^{m_3}}{d_{50}^{m_4}} \end{cases} \quad (9)$$

where  $A, B, A^*, B^*, n_1$  to  $n_3, m_1$  to  $m_4$  are fitting constants.  $d_{50}$  is regarded as the surface average diameter.  $I_C, f_C,$  and  $K_B$  represent the inter-particle cohesion, frequency of Brownian collisions and nucleation rate constant of cement suspensions, respectively.  $S_d$  is the simplified average separation distance between cement particles, which can be calculated as:

$$S_d = d \cdot \left[ (\phi / \phi_m)^{-1/3} - 1 \right] \quad (10)$$

where  $d$  is the mean particle diameter from BET specific surface area measurements, as expressed by  $d = 6 / (\text{BET} \times \text{density of cement})$ ,  $\phi$  is the solid volume fraction, and  $\phi_m$  is the maximum volume fraction. Detailed introductions and calculations of these parameters are referred to Ref. [21]. The proposed model quantitatively describes the physico-chemical kinetics of structural build-up of neat cement suspensions.

It should be mentioned that different from the static yield stress, the critical strain corresponding to the limit strain of the LVER indicates the strength of connections of early hydration products between cement particles [16,60]. Similar evolutions of static yield stress and storage modulus are generally obtained [23]. However, quite different trends of evolution of structural build-up of cement paste can also be concluded by stress growth test and SAOS test. For example, Ma et al. [61] stated that the addition of diutan gum increased inter-particle distance and attractive interactions between networks due to the interpenetration and entanglement of absorbed diutan gum molecules on cement particles, resulting in a decrease in the interparticle links and an increase in the flocculation structures. This led to a rapid increase in static yield stress and slow evolution of storage modulus for the same paste mixture. Since the different origins of various evaluation methods, the combination of static yield stress and SAOS test will be accordingly beneficial to monitor the structural build-up of cementitious materials more accurately [62].

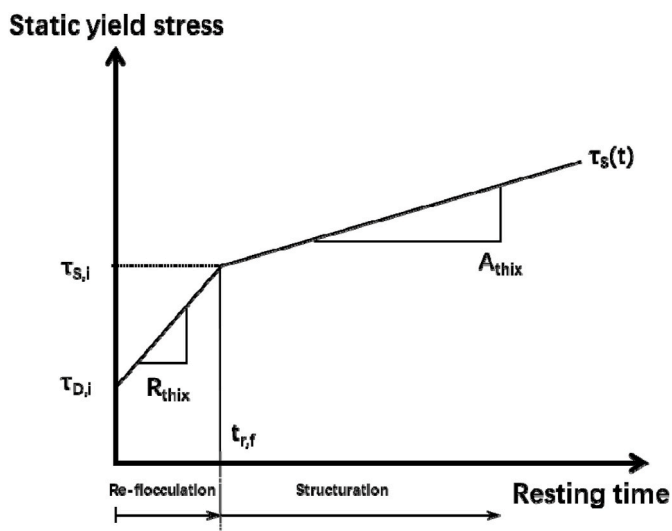


Fig. 3. Bi-linear model of static yield stress describing structural build-up (adapted from Ref. [53]).

### 3.4. Conventional workability tests

Penetration tools like Vicat needles and penetrometers are often used to measure the static yield stress of cementitious materials in the case of lack of suitable rheometric measurement methods for very stiff suspensions. The theoretical base of this method is according to the fact that the yield stress of the material exerts a resistance to the penetrating needle. The stress created by the penetrating needle is dependent on the shape of the needle as well as the applying speed. Considering the penetrating velocity is sufficiently low, the induced flow can be regarded as quasi-static flow. Based on this assumption, Lootens et al. [63] established the correlations between the penetration test parameters and the yield stress of the material, as summarized in Table 2. Although the penetration tests are not sensitive to the early changes of the microstructure, it can be widely used to characterize the structural build-up of cementitious materials [64,65]. For example, Ma et al. [66] found that the penetration resistance showed an approximately linear relationship with resting time for 3D printing stiff concrete materials. Yuan et al. [67] stated that the growth rate of penetration resistance had a linear relationship with the growth rate of static yield stress. Therefore, the penetration tests could be an alternative method for evaluating the structural build-up of cementitious materials, especially for those with highly thixotropic properties.

As aforementioned, the flow induced by the penetrometer is assumed to be quasi-static. Amziane et al. [68] developed a simple device with a perfectly static rough plate immersed in a suspension to monitor the evolution of static yield stress with time. The schematic diagram of the plate device and the corresponding principle are presented in Fig. 4. The theoretical hypothesis of this device is based on the fact that a measurable stress is generated by the deformation of the suspension, such as sedimentation and time evolution of yield stress. For a homogeneous cementitious paste, the evolution of shear stress with time can be calculated by monitoring the mass variation of the plate surface:

$$\tau(t) = \frac{g}{2} \left[ \frac{\Delta M(t) + \alpha(T) \cdot t}{l \cdot H} - e \cdot \rho \right] \quad (11)$$

$$\Delta M(t) = -l \cdot e \cdot \int_0^H \rho(z \cdot t) dz + \frac{l}{g} \cdot \int_0^H 2\tau(z \cdot t) dz - \alpha(T) \cdot t \quad (12)$$

where  $l$  and  $e$  are respectively the length and thickness of the plate,  $H$  is the height of the plate immersed in the suspension,  $\alpha(T)$  is a temperature-dependent parameter,  $\rho$  is the local density of the suspension,  $\tau$  is the local shear stress,  $t$  is resting time, and  $\Delta M$  is the apparent mass variation of the plate. Results from a cement paste with  $w/c$  of 0.35 show that the shear stress measured by the plate shows a comparable evolution to the yield stress determined by rheometer. This method can be used to evaluate the structural build-up of a suspension with obvious deformation [68,69]. Furthermore, the principle of this method has been extended to design a static Vicat test, which can be used to monitor the evolution of yield stress of cement paste immediately after mixing

**Table 2**  
Relations between penetration test parameters and yield stress [63].

Test geometries	Measurement type	Relations
Vicat needle with radius $R$	Penetration depth $h$ under load of 300 g	$\tau_0 = \frac{3}{2\pi Rh}$
Hemispherical penetrometer with radius $R$	Force $F$ under given speed	$\tau_0 = \frac{F}{3\pi R^2}$
Hemispherical penetrometer with radius $R$ connecting a cylinder with height $h$	Force $F$ under given speed	$\tau_0 = \frac{F}{3\pi R^2 + 2\pi Rh}$
Conical penetrometer with radius $R$ and height $h_2$ connecting a cylinder with height $h$	Force $F$ under given speed	$\tau_0 = \frac{F}{\pi R \sqrt{R^2 + h_2^2} + 2\pi Rh}$

[70,71].

Khayat et al. [72] proposed an inclined plane method as a simple approach to evaluate the structural build-up with time for high flowable cement-based materials such as SCC. The schematic diagram of this device is shown in Fig. 5. Under the combination of gravitational force and frictional force, the sample on the inclined plane starts to flow downward when the angle  $\alpha$  reaches a particular critical angle  $\theta$ . The correlation between the critical angle and the static yield stress ( $\tau_0$ ) of the material can be expressed as:

$$\tau_0 = \rho g h_e \sin \theta \quad (13)$$

where  $\rho$  is the density of the material,  $g$  is the acceleration gravity, and  $h_e$  is the effective height. The critical angle indicates the intensity of the structural build-up, with higher critical angle meaning stronger internal microstructure. Note that the inclined plane needs to be coated by superfine sandpaper with grit number of 600, necessitating changes every 10 to 15 tests. The authors found that the evolution of structural build-up of flowable concrete with slump flow ranging from 560 mm to 720 mm determined by the inclined plane test showed excellent correlation with that obtained using rheometer. Furthermore, the same authors also developed some other field-oriented methods such as portable vane, undisturbed slump spread, cone penetration and K-slump to evaluate the structural build-up of SCC [73]. It is found that the cone penetration test could successfully monitor the structural build-up of concrete. Similarly, Jarny et al. [74] also performed inclined plane tests to determine thixotropic parameters. Recently, Megid and Khayat [75] stated that the thixotropic build-up of SCC had a strong correlation with the workability loss indexes obtained from slump and J-ring flow test methods. In other words, the structural build-up of SCC to an extent can be assessed by the conventional workability test methods.

### 4. Influencing factors

The thixotropic structural build-up is a combined result of colloidal interactions and chemical hydration. Based on the origins of structural build-up, Lowke [38] established a quantitative model to correlate the structural build-up rate of SCC with surface coverage of superplasticizer and particle packing. The surface coverage affects the particle separation, and thus influences the colloidal interactions and hydration reactions, while the effect of particle packing on the structural build-up can be explained by the particle contacts and crowding effects. The surface coverage indicates the attraction between two particle surfaces, referring to the colloidal surface interactions, whereas the particle packing describes the contact interactions between particles. Based on the YODEL model of suspensions [76,77], the relationship between the structural build-up, the particle packing and the surface coverage can be expressed as:

$$A_{thix} = m \cdot \frac{1}{\theta^{3.2}} \cdot \frac{\phi^3}{\phi_m \cdot (\phi_m - \phi)} - n \quad (14)$$

where  $\theta$  is the surface coverage which can be calculated from isotherm [38],  $\phi$  and  $\phi_m$  are the particle volume fraction and maximum packing fraction, respectively, and  $m$  and  $n$  are fitting constants. Eq. (14) provides a quantitative and fundamental theoretical relation between the structural build-up and physical parameters of the suspension. It can be seen that the structural build-up has a positive association with the particle volume fraction, while opposite correlations with the surface coverage and the maximum packing fraction, due to the fact that crowding effects and particle contacts increase with particle volume fraction but decrease with increasing maximum packing density [38]. That is, the factors influencing the surface coverage and particle packing of the cementitious systems play significant roles in the thixotropic structural build-up. In this section, the influences of constituents, mixture proportions and external factors such as temperature and magnetic field on the thixotropic structural build-up of fresh



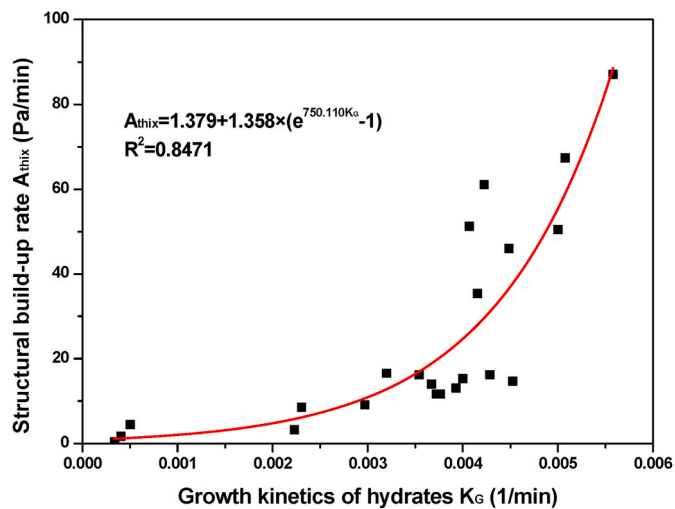


Fig. 7. Relationship between structural build-up rate and growth kinetics of hydrates (adapted from Ref. [83]).

cement particles (such as fineness) in favor of increasing particle density and accelerating chemical hydration also exert positive effects on the improvement of structural build-up of cementitious materials.

The influence of supplementary cementitious materials (SCMs) on the structural build-up depends on their particle size distribution, particle shape and roughness. The addition of fine and/or irregular SCMs such as slag, silica fume and metakaolin increases the structural build-up [50,59,81,85,86], possibly due to the increase in particle volume fraction and the pozzolanic reaction. On the one side, the incorporation of SCMs gives a rise to the particle volume fraction and internal frictional forces between solid grains, resulting in a higher flocculation rate. On the other side, the cation precipitation by the formation of calcium hydroxide as a precursor of the pozzolanic reaction of SCMs creates more links and bridges between particles [86]. The degree of the change of structural build-up is dependent on the physical properties and chemical compositions of SCMs, as well as the w/b ratio. For example, Yuan et al. [49] stated that silica fume increased the growth rate of static yield stress, whereas ground slag showed an opposite response. Ferron et al. [87] found that the influence of silica fume on the structural build-up depended on the w/b ratio, with no significant change at w/b of 0.35 and an apparent increase at w/b of 0.45, respectively. Moreover, Navarrete et al. [81] stated that the particle size of SCMs played its role in the structural build-up of cementitious materials through particle density (by controlling the particle contacts) rather than specific surface area, and the surface potential of SCMs showed a more significant effect on the structural build-up than the chemical reactivity, with the effect lower than that of the particle density.

In the case of fly ash, many studies [49,87,88] stated that fly ash particles slowed down the structural build-up of cement pastes. From the physical viewpoint, spherical fly ash particles with micro-filling effect decrease the water demand and then more free water can be created, contributing to higher thickness of water layer and thus higher interparticle distance. From the chemical aspect, the rate of hydration of the cementitious system is postponed with the replacement of cement with fly ash, prolonging the dormant period [89,90]. Accordingly, fly ash particles generally reduce the rebuilding potential of cement pastes. Nevertheless, the opposite result that the addition of fly ash can exert an increase in the structural build-up of cement-based materials is often reported by researchers. Ahari et al. [51] stated that the use of fly ash increased the structural build-up of SCC, irrespective of class C and F fly ashes. Rahman et al. [85] found that increasing fly ash content from 5% to 10% resulted in a significant increase in flocculation rate, and the structural build-up rate of mixture with 10% fly ash was 3.5 times higher than that of reference concrete. This behavior can be attributed to the

increase in the particle contacts with increasing solid volume in the mixtures.

Limestone powder is a popular inert filler in cementitious materials. In the case of fine ground limestone powder, it has lower average particle size and higher specific surface area than cement particles [91]. The replacement of cement with limestone powder by mass increases the solid concentration, reducing the particle surface distance. Accordingly, the substitution of limestone powder is beneficial to increase the structural build-up of cementitious materials [85]. Nevertheless, Zhang et al. [92] stated that the addition of limestone powder reduced the cohesive forces due to ion-ion connections and the hydration kinetics, and therefore decreased the structural build-up rate of cement paste. Due to the opposite effect of fineness of limestone powder on the interparticle forces and hydration kinetics compared to the concentration, increasing the fineness of the limestone powder showed an increase in the structural build-up rate [93].

#### 4.3. Superplasticizer

In the presence of Polycarboxylate Ether (PCE) superplasticizer, steric repulsive forces are introduced, which dominate the electrostatic double layer repulsion forces [94,95]. This leads to an increase in the distance between particle surfaces and effective layer thickness, which decreases the colloidal interactions and increases the bridging distances between cement particles [76,96]. Consequently, the addition of superplasticizer could exert a significant influence on the thixotropic structural build-up of cement-based materials [16,38,97]. The influence of superplasticizer on the structural build-up relies on the type, structure, dosage, w/b, cementitious composition, as well as the adsorption and the degree of surface coverage [38,87,98–100].

Qian et al. [101] stated that both adsorbed part and remaining part in the interstitial pore solution increased with the dosage of PCE superplasticizer. They also found that lower PCE additions dramatically decrease the thixotropic structural build-up, and the value remains low and unchanged with PCE dosages higher than the Critical Micelle Concentration (CMC). The CMC corresponds to the concentration of surfactants with the first single layer adsorbing on cement particles, changing them from hydrophilic to hydrophobic [102]. Below the CMC, the surface of cement particles is not fully covered by PCE molecules, and thus the cement paste tends to be more agglomerated. Beyond the CMC, however, the adsorption of PCE surfactants is saturated. In this case, the bridges of early hydration products might be covered by PCE molecules, or only exist in the connections between single cement particles within agglomerates. This results in weak colloidal bonding between agglomerates, and thus low thixotropic structural build-up [101]. The saturation dosage differs for different superplasticizers, for example, with PCE saturation dosage of 0.1% while Naphthalene Sulfonate Formaldehyde (NSF) saturation dosage of 0.6% [103]. This can be attributed to the adsorption mechanisms, where the adsorption model of NSF is based on a monolayer model, while PCE follows a multi-layer adsorption [104].

The structure of superplasticizer affects the thixotropic behavior. Nicia and Lowke [99] observed that reducing PCE side chain density and length increased the thixotropic structural build-up of cementitious materials. This can be explained by the adsorption behavior of PCE molecules. PCE molecules with low side chain densities have a faster adsorption speed. Accordingly, less PCE molecules might be available in the interstitial solution to adsorb on newly formed hydration products, resulting in a decrease in steric repulsion and thus an increase in the thixotropic structural build-up. PCE molecules with short side chain length decrease the PCE layer thickness on the surface of cement particles, having a potential to be overgrown by new hydration products.

There are generally two approaches to obtain a desired flowability. One is increasing w/b and the other is adding superplasticizer while keeping low w/b. Ferron et al. [87] found that the second approach could significantly increase the rate of rebuilding of SCC. The authors



also stated that at low flowability, PCE-based superplasticizer showed a slightly higher enhancement in structural build-up rate than the NSF, whereas an opposite behavior was observed at relatively high flowability.

#### 4.4. Viscosity-modifying agent

Viscosity-modifying agent (VMA) is widely used to trigger the viscosity and flocculation rate of fresh concrete mixtures. According to the chemical compositions, VMAs are organic or inorganic. Typical organic VMAs include cellulose ether and Welan gum. Nano-silica and nano-clay are two kinds of representative inorganic VMAs.

The addition of organic VMA considerably increases the thixotropic structural build-up of fresh cement-based materials. On the one side, the long polymer chains in VMA molecules have a potential to physically adsorb water, reducing the free water in the interstitial solution. On the other side, attractive forces develop between the VMA molecules, facilitating the formation of gel structure and inter-particle links. A higher VMA concentration corresponds to a greater effect. As a result, the thixotropic structural build-up of cement-based materials increases with the content of VMA [50]. Furthermore, the efficiency of VMA depends on the structure and the used superplasticizer. Assaad and Khayat [105] stated that compared to SCC mixtures with powder or liquid polysaccharide-based VMA and NS-based superplasticizer, SCC mixtures containing cellulose-based VMA along with PCE-based superplasticizer exhibited higher initial structural breakdown area but lower growth rate of thixotropic structural build-up. The lower structural build-up rate is probably due to the improvement in flowability retention by the PCE-based superplasticizer, which reduces the shear stress measured after longer period and thus leading to lower rate of increase in thixotropy [105].

Inorganic VMAs such as nano-silica and nano-clay attract great attention with the advancement of 3D concrete printing. Quanji et al. [106] found that the optimum nano-clay content was around 1.3% by mass of cement to reach a peak in the rate of structural rebuilding. Yuan et al. [49] pointed out that nano-silica is the most effective mineral additive to improve the structural build-up of cement paste among ground slag, silica fume, attapulgite and nano-calcium carbonate. The increase in structural build-up can be attributed to the nucleation effect and micro-filling effect of ultra-fine particles. The oppositely charged edges of nano-clay have potential to associate together by electrical attraction forces, forming denser internal microstructures. Moreover, the van der Waals forces in the solid system generally increase due to the smaller particle size of nano-clay, showing a positive impact on the thixotropic structural build-up [107]. The increased effect of nano-clay on the structural build-up can be more pronounced when the superplasticizer is in an appropriate dosage [62]. Interestingly, the addition of nano-clay indeed has an immediate increasing effect on the initial static yield stress. However, it seems that the nano-clay shows little influence on the structural build-up rate [61,108]. This is probably due to the fact that the rigidification rate is dominated by the chemical hydration of cement particles [109].

#### 4.5. Aggregates and rigid fibers

Mahaut et al. [110] stated that the thixotropic structural build-up of fresh concrete is independent of the presence of solid particles. In other words, the origin of structural build-up of concrete is mainly attributed to the cement paste. The structuration rate of fresh concrete can be predicted from the structural build-up rate of cement paste, which can be expressed by Eq. (15).

$$A_{thix,conc} = A_{thix,paste} \sqrt{\frac{1-\phi}{(1-\phi/\phi_m)^{2.5\phi_m}}} \quad (15)$$

where  $A_{thix,conc}$  and  $A_{thix,paste}$  are respectively the structural build-up

rate of fresh concrete and corresponding cement paste,  $\phi$  and  $\phi_m$  are the aggregate volume fraction and maximum aggregate packing fraction, respectively. Lecompte et al. [111] experimentally verified this model, and found that it can successfully predict the structural build-up of cementitious materials at solid packing fraction within the random loose packing volume fraction. In other words, the concrete rheology is mainly controlled by thixotropy below the random loose packing fraction, whereas at relatively high solid volume fractions, the concrete rheology is mainly governed by frictional aggregate interactions.

Irrespective of the relations to the cement paste, the thixotropic structural build-up of concrete is affected by the volume and characteristics of aggregates. Omran et al. [112] stated that increasing aggregate volume (inversely decreasing the paste volume) increases the thixotropic structural build-up of SCC. This is in good agreement with the findings of Assaad and Khayat [113]. This is due to the higher internal friction and particle interlocking effect. Under the same paste content, the structural build-up of SCC increases with the decrease of sand to total aggregate ratio. In the case of aggregate size, higher thixotropy corresponds to coarse aggregate with larger nominal maximum size. In spite of the inert nature of aggregates, the experimental results obtained by Ivanova and Mechtcherine [114] allow to pose a hypothesis that there might exist physical-chemical interactions between fine aggregates and cement paste. Further research should be conducted to verify this hypothesis and a proper model should be proposed to characterize the effect of aggregates on the structural build-up of cement-based materials.

The structural build-up of cementitious materials containing rigid fibers can also be evaluated by using quantitative models. Perrot et al. [115] extended the model of Mahaut et al. to rigid fiber reinforced cement-based materials, which can be written as follows:

$$A_{thix,conc}(\phi_s, \phi_f) = A_{thix,paste} \sqrt{\frac{1-\phi_m P_f}{(1-P_f/0.8)^{2.5-P_f/0.8}}} \quad (16)$$

where  $A_{thix,conc}(\phi_s, \phi_f)$  is the structural build-up rate of fresh concrete containing rigid fibers.  $\phi_m$  is the dense packing fraction of the sand, and  $P_f$  is the relative volume fraction of the inclusions [116], which can be calculated by:

$$P_f = \phi_f \cdot \frac{r}{4} + \frac{\phi_s}{\phi_m} \quad (17)$$

where  $\phi_f$  is the volume fraction of fibers and  $4/r$  is the dense packing of fibers.  $\phi_s$  is the volume fraction of sand. The modified model shows that the structural build-up of rigid fiber reinforced cement-based materials only depends on the relative volume fraction. The experimental results show that at the relative volume fraction lower than the random loose packing fraction, the structural build-up of composite materials can be perfectly predicted by Eq. (16), while at high relative volume fractions, the predicted structural build-up is larger than the actual one because the strong inclusion network reduces the effect of cement paste structuration.

#### 4.6. Temperature

The ambient temperature has a significant influence on the colloidal interactions and hydration kinetics, and thus the thixotropic structural build-up of cement-based materials. Increasing temperature increases the dissolution rate of anhydrous clinker phases [117,118]. From the viewpoint of physical effect, on the one hand, the increase of ionic concentration and the production of a large number of fine particles improve the attractive interactions between solid particles. On the other hand, the Brownian motion increases with elevated temperature. Consequently, the flocculation rate of cement suspensions increases with the temperature. From the chemical viewpoint, the hydration rate of cementitious materials increases with the elevated temperature, and

thus more hydration products will be generated. The increase of number and size of bridges of early hydration products between cement particles imposes a positive effect on the improvement of thixotropic structural build-up. Therefore, higher temperature generally leads to faster rate of structural build-up of fresh cementitious materials. Nevertheless, Bogner et al. [44] observed a temperature-independent increase in shear modulus within 1.4 h after the first contact of water and cement particles, possibly due to the similar phase deposition on the surface of cement particles.

The influence of temperature on the structural build-up depends on the cementitious compositions of the material and the presence of chemical admixtures. Huang et al. [119] evaluated various mineral additions on the temperature-dependent structural build-up, as depicted in Fig. 8. It can be found that the variations of structural build-up rate of the cementitious pastes with increasing temperature from 20 °C to 40 °C were much larger than that obtained with temperature increasing from 10 °C to 20 °C. It is also revealed that compared to the neat cement paste, fly ash, slag and silica fume showed a negative effect on the structural build-up at 10 °C, while a positive influence was obtained at 40 °C. In contrast, the incorporation of nano-CaCO<sub>3</sub> and nano-silica was favorable to the structural build-up of cementitious paste, regardless of the temperature. Vanhove et al. [120] stated that SCC mixtures containing both NSF-based superplasticizer and polysaccharide-based VMA had the lowest thixotropic structural build-up at temperature of 21 °C, compared to other temperature conditions. At relatively low temperatures, the plasticizing effect of PCE is insignificant, resulting in dominating attractive forces between solid particles and thus higher thixotropic structural build-up. At temperature higher than 21 °C, however, less PCE molecules existing in the interstitial solution lead to an increase in the coagulation of cement particles.

#### 4.7. Magnetic field

External trigger signals such as magnetic field could exert a significant influence on the structural build-up of cementitious materials with magnetizable particles. Nair and Ferron [121,122] confirmed that the evolution of storage modulus of fresh cement paste containing carbonyl iron particles did show field sensitivity to an external magnetic field. This can provide an effective approach to control the structural evolution of cement-based materials on-demand.

Recently, De Schutter and his co-authors [60,123–125] investigated the influence of magnetic field on the structural evolution of cementitious paste containing magnetizable particles or advanced polymers. A

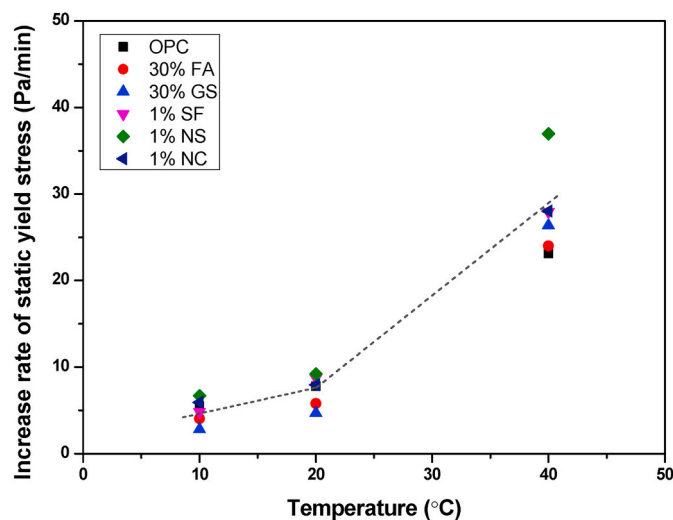


Fig. 8. Effect of temperature on the structural build-up rate of cementitious paste (Data derived from Ref. [119]).

typical experimental result of nano-Fe<sub>3</sub>O<sub>4</sub> containing cement paste under step-changed magnetic field is shown in Fig. 9. It can be found that suddenly applying a magnetic field to a resting cementitious paste results in a drop reduction in the storage modulus, and afterwards the storage modulus increases significantly. The drop reduction of the storage modulus can be attributed to the micro-agitation effect of moving nanoparticles under magnetic field, and the significant increase is due to the formation of chains or clusters structures [126–128]. Despite requiring more in-depth research, it can be concluded that the structural build-up of cementitious paste with magnetizable particles can be controlled on-demand by using magnetic field.

Overall, the influence of constituents, mixture proportions and external factors on the thixotropic structural build-up of fresh cement-based materials can be summarized in Table 3.

## 5. Applications of structural build-up in cementitious materials

### 5.1. Formwork pressure

Formwork pressure of concrete is of great interest in construction because it affects construction cost, speed and safety. With the growing application of flowable concrete and SCC, it is important to predict the formwork pressure. Under sufficiently low casting rate or at rest state, cementitious materials can build up internal structures. Together with the internal frictions between solid particles, the concrete in the bottom of formwork has the ability to withstand the load from the above concrete. This results in a reduction in the lateral stress exerted to the formwork with the value much lower than hydrostatic one [129–131]. Assuming that the casting rate is constant and the shear stress at the wall exerted by the vertical deformation of the concrete under gravity is equal to the yield stress, the relative formwork pressure can be predicted by Eq. (18) [19,132].

$$\sigma^*(z) = \frac{\sigma_{xx}}{\rho g H} = 1 - \frac{H A_{thix}}{\rho g e R} \quad (18)$$

where  $\sigma^*(z)$  is the relative lateral pressure,  $\sigma_{xx}$  and  $\rho g H$  are the lateral stress and the associated hydrostatic pressure, respectively,  $R$  is the casting rate,  $H$  is the depth of the concrete, and  $e$  is the thickness of the concrete. It should be noted that this model is applied to common rectangular formwork with length and height far larger than width. According to Eq. (18), we can see that the lateral stress is equal to the hydrostatic pressure if casting at a high speed or for concrete with low flocculation rate. However, for the concrete with high thixotropic

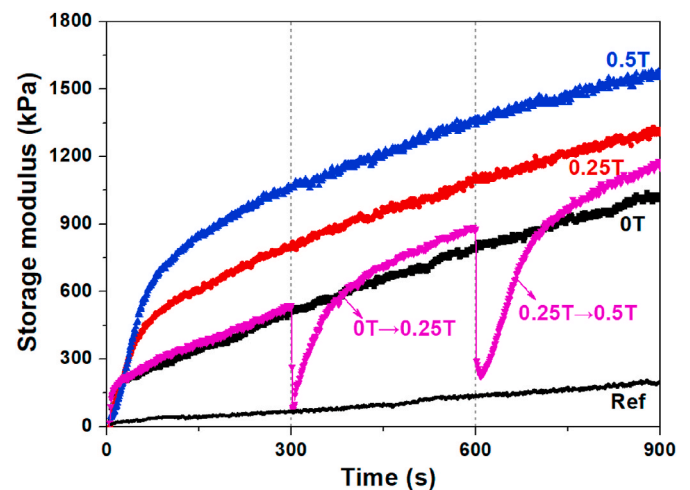


Fig. 9. Evolution of storage modulus of cementitious paste (w/c of 0.4 and nano-Fe<sub>3</sub>O<sub>4</sub> of 5%) under step-increased magnetic field (adapted from Ref. [60]).

**Table 3**  
Summary of influencing factors of thixotropic structural build-up.

Factors	Effect on structural build-up
Increase w/b	Decrease
Fly ash	Decrease (rarely increase)
Slag, silica fume, metakaolin	Increase
Superplasticizer	Decrease
VMA (cellulose ether, nano-clay, etc.)	Increase
Accelerator	Increase
Retarder	Decrease
Aggregates or stiff fibers	Increase
Elevated temperature	Increase
Magnetic field	Control on-demand (magnetizable particles are required)

structural build-up and casting at low rate, the lateral stress is far lower than the hydrostatic pressure. This physical model provides a quantitative correlation between the lateral stress and the thixotropic structural build-up.

However, since the model in Eq. (18) is based on the assumption that the static yield stress linearly evolves over time, the predicted lateral pressure might be overestimated for a longer period (e.g., longer than 1 h). In this case, Perrot et al. [133] extended the model considering the exponential evolution of yield stress with time (as illustrated in Eq. (4)). For a common rectangular formwork with the length far larger than the width, the extended model describing the relative lateral pressure can be written as:

$$\sigma^*(z) = 1 - \frac{2R}{\rho g e H} \left[ A_{thix} t_c \left( t_c \left( e^{\frac{H-e}{R t_c}} - 1 \right) - \frac{H-e}{R} \right) \right] \quad (19)$$

where  $t_c$  is a characteristic time to obtain the best fit with experimental values. At higher casting rates, both Eqs. (18) and (19) can give similar predictions to the measured lateral pressure. However, for lower casting rates with casting time longer than a critical time (usually can be regarded as 1 h), Eq. (19) gives a more accurate relative lateral pressure than Eq. (18).

Omran and Khayat [134] established the relationships between the formwork pressure characteristics (including maximum pressure after casting and pressure decay) and structural build-up indices. In the case of the relative maximum lateral pressure at various casting depths, defined as the ratio of actual lateral pressure and equivalent hydrostatic pressure, it has a strong correlation with the structural build-up indices considering the coupled effect of structural build-up at 15 min and corresponding change with time determined by rheometer or portable vane. The pressure decay at the first hour after casting is well correlated with the viscosity difference between the initial and the equilibrium determined by a rheometer, while the indices considering the coupled effect of structural build-up at 15 min and corresponding change with time determined by rheometer or portable vane are recommended to describe the total pressure decay. Furthermore, by conducting a series of round-robin tests, Billberg et al. [135] evaluated ten different models considering different parameters (e.g., structural build-up, slump loss, setting time, etc.) to predict the lateral pressure. The authors concluded that all the models can provide lateral pressure predictions with sufficient accuracy. More detailed information is referred to Ref. [135].

## 5.2. Multi-layer casting

Since SCC generally has high thixotropic structural build-up, during the placement, the apparent yield stress of the casting layer may increase to a high value, resulting in a not intermixed new layer [136]. This behavior induces lift lines and weak interfaces between two casting layers [137], showing a negative influence on the concrete quality and structural durability [138]. Roussel and Cussigh [136] established the relationship between the apparent yield stress, the flocculation rate and

the interval time between two casting layers, as described by Eq. (20).

$$\tau_0(\Delta t) = \tau_0 + A_{thix} \cdot \Delta t \quad (20)$$

where  $\tau_0$  and  $\tau_0(\Delta t)$  are the initial yield stress and the yield stress after a resting period  $\Delta t$ , respectively. A critical interval time after which two layers with thickness of  $h$  will be not mixed well can be defined as:

$$\Delta t_c = \frac{\rho g h}{2\sqrt{3}A_{thix}} \quad (21)$$

Based on Eq. (21), the critical interval time can be used as a parameter to guide the construction process. For example, for a SCC mixture with structuration rate of 0.3–0.5 Pa/s and the thickness of the second layer higher than 10 cm, the critical interval time is about 20–30 min. This means that the concrete quality will be not significantly influenced when casting the multi-layer concrete within 20–30 min.

Besides, Megid and Khayat [137] correlated the bond strength of successive layers to the structural build-up of SCC mixtures by an empirical method. Take direct shear strength as an example, the critical interval time corresponding to 90% of residual bond strength can be estimated by Eq. (22).

$$\Delta t_c = 1072 - 10.72 R_{Dsh}\% + (1.36 R_{Dsh}\% - 136) LnPAI \quad (22)$$

where  $R_{Dsh}$  is the residual direct shear strength (%), and PAI is the passing ability index from a J-ring test (mm.mm/min) characterizing the structural build-up of SCC. Experimental results show that SCC mixtures securing high residual bond strengths should have relatively low structural build-up with the maximum PAI value lower than 600 mm mm/min [137].

## 5.3. 3D concrete printing

In extrusion based additive manufacturing, e.g., 3D concrete printing, the concrete is generally extruded layer by layer. The interval time between two extrusion layers has to be optimized to reach a balance between long period to obtain adequate mechanical strength for the bottom layer and short enough to obtain a sufficient bonding strength between two layers [139–142]. The mechanical strength of each layer, mainly from the yield stress of the material, enables to keep the stability under its own weight and the load from upper layers. A higher yield stress corresponds to a higher buildability and a lower deformation of the bottom layer. However, the material cannot possess a high yield stress at the beginning, as it has to be sufficiently fluid for pumping and extrusion. In this context, the requirements of rheological properties for 3D printing concrete are summarized in Ref. [143]. Besides, controlling the structural build-up of cement-based material is another potential solution to ensure sufficient fluidity during extrusion and high micro-structural strength evolution after extrusion.

Generally, fast building rate is beneficial to 3D extrusion printing. In other words, longer interval time and higher thixotropy indicate weaker interface connection. By compressing a specific concrete mixture with various construction rates, Perrot et al. [47] proposed a theoretical model to correlate structural failure during 3D printing with the structural build-up. Instead of a single layer, the authors used a cylinder as an extruded structure. For a circular column layer with diameter of  $D$ , considering the structural build-up of the concrete following the linear evolution model provided by Roussel [19], the proposed failure time can be described by Eq. (23).

$$t_c = \frac{\tau_0}{\rho g R / \alpha_{geom} - A_{thix}} \quad (23)$$

where  $t_c$  is the critical failure time, after which the extruded layer is expected to collapse,  $\tau_0$  is the initial static yield stress,  $R$  is the construction rate,  $\alpha_{geom}$  is a geometric parameter depending on the shape of the printed structure. For the circular column layer, this parameter can

be calculated by:

$$\alpha_{geom} = 2 \left( 1 + \frac{D}{2\sqrt{3}h} \right) \quad (24)$$

where  $h$  is the height of the column layer. The experimental results of the vertical stress, failure point and predicted critical stress are plotted in Fig. 10. It can be seen that the predicted critical failure times were in good agreement with the experimental observations for a 70 mm diameter column under construction rates varying from 1.1 to 6.2 m/h [47]. It should be mentioned that Eq. (23) seems to contradict the fact that higher structuration rate suggests shorter open time. More experiments should be performed to establish the relationship between failure time and structuration rate of concrete mixtures.

Recently, Roussel [143] stated that high structural build-up rate could result in cold joints. Cold joints often occur between two connecting layers beyond a critical time. From the structural build-up point of view, the maximum interval time can be derived, as can be described by Eq. (25).

$$T_{max} = \frac{\sqrt{\frac{(\rho g h_0)^2}{12} + \left(\frac{2\mu_0 V}{h_0}\right)^2}}{A_{thix}} \quad (25)$$

where  $\mu_0$  is the plastic viscosity of the printed material,  $\rho$  is the density of the printed material,  $h_0$  is the layer thickness, and  $V$  is the nozzle velocity. This relation provides a useful method to predict the maximum time for a single layer to be produced.

#### 5.4. Numerical thixotropy modelling

Another important application of thixotropy in general or build-up of cementitious materials is the prediction by numerical simulation. Thixotropy has a significant effect on the formwork pressure decay, inter-layer bonding of concrete casting or 3D printing, pumping re-initiation after (un)expected rest and many more [10,26,29]. Despite the importance of thixotropy on several aspects of fresh concrete simulation, only a handful of simulations did consider thixotropy. Most of these simulations were performed with a homogeneous type of computational fluid dynamics (CFD) software, either considering a finite volume or volume of fluid type of method. A literary overview is outlined.

Roussel [136] simulated the influence of thixotropic build-up on the

intermixing of delayed casting layers during the concrete casting process. Several numerical simulations with a different resting period in between consecutive casting layers were performed with commercial CFD software FLOW3D®. Two cases were elucidated in particular. The first one illustrated a layer casting delay of 300 s, with a yield stress of 50 Pa, a plastic viscosity of 50 Pa.s, a thixotropic build-up of 0.5 Pa/s and a de-flocculation rate of 0.005. The second one considered a casting delay of 1200 s. On basis of the thixotropic simulations, the authors concluded that a clear distinction could be made when two layers were intermixed and when not. This was concluded from whether the generated stresses were sufficient to initiate flow in the previous layer or not. For the shortest casting delay of 300 s, the layers were well intermixed resulting in a smooth interface between intermixed layers. For the longest casting delay of 1200 s, it resulted in a sharp interface between consecutive layers. The imposed stresses of the latter were not sufficient to cause flow initiation and thereby insufficient intermixing of the layers.

Another example is the thixotropy simulation in CFD software Viscometric-ViscoPlastic-Flow® developed by Wallevik J.E [144], where the Couette flow geometry of the ConTec Viscometer 4 was simulated. Experimental rheological results were compared with the simulations of the fitted Hattori-Izumi model, modified Hattori-Izumi model (after [144]) and other models. Unlike the other thixotropy models presented by Ref. [144], it was concluded that the rheometer torque response was well in agreement with the fitted modified Hattori-Izumi model. Similarly [22], concluded that the extended Hattori-Izumi model can explain the transient phenomena commonly observed for cement pastes. Later on, Wallevik [15] further refined former modified Hattori-Izumi (MHI) theory and respectively named it the Particle Flow Interaction theory with version mark I (PFI-I). The PFI theory was later improved with regard to structural breakdown-components, resulting in mark version II (PFI-II) [15]. Some additional thixotropy simulations have been performed by the respective author using the PFI-II theory - implemented in open source CFD software OpenFOAM - in some conference proceedings such as the simulation of casting of T-beam [145]. Despite the fact that a good transient thixotropic behavior could be simulated and the PFI theory stems from a fundamental kinetic approach, numerous model parameters are required and cannot straightforwardly be determined.

Thixotropy has also been modelled in the simulation of pipe flow of cementitious suspensions [146]. The effect of thixotropic build-up of cementitious suspensions in pipelines was also briefly numerically

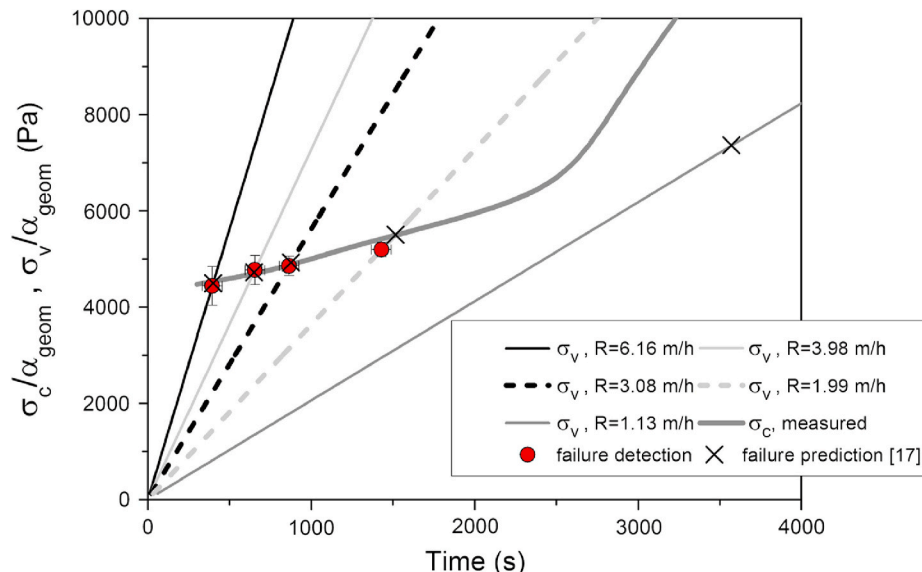


Fig. 10. Comparison between the critical stress and vertical stress [47].

investigated by Ref. [147]. In former works, the simplified thixotropy model of Roussel [19] was implemented in OpenFOAM. Despite some successful simulations, some numerical stability issues are not fully addressed. Lastly, Tan et al. [148] modelled thixotropic concrete in a discrete element method (DEM), by considering a time-dependent contact parameter. A numerical investigation on formwork wall pressure was performed. After a model calibration based on an L-box and rheometer test, the numerical results were in accordance with the experimental observations, demonstrating their model capability to some extent.

### 5.5. Active stiffening control

Contradicting requirements of fresh properties exist in different casting operations of cement-based materials. For example, high structural build-up and low flowability are beneficial to reduce formwork pressure, as can be easily observed from Eq. (18). By contrast, low structural build-up and high flowability are favorable to the multi-layer casting process (shown as Eq. (21)). This means that it is necessary to strike a balance between the opposing property requirements in different casting processes for the same concrete mixture. This highlights the importance of rheology and stiffening control [149,150]. On this issue, De Schutter proposed an advanced concept, i.e., “SmartCast” [151,152], aiming at controlling the rheology and stiffening of cement-based materials by using external trigger signals.

From the viewpoint of active structural build-up control, one potential approach is incorporating magnetic nanoparticles and applying a magnetic field, with a representative example presenting in Fig. 11. The responsive cementitious paste with w/c of 0.45 and nano-Fe<sub>3</sub>O<sub>4</sub> (200 nm) content of 3% (by mass of cement paste) has comparable structural build-up to the reference cement paste with w/c of 0.4. After applying a constant magnetic field with 0.5 T, the storage modulus of the responsive mixture significantly increases, and its value at 90 min after applying the magnetic field is 1.5 times larger than that obtained without magnetic field. This implies the significant increase in the stiffness of the cementitious paste due to the formation of magnetic clusters. When a pulsed magnetic field lasting for 10 s is applied to the responsive mixture every 10 min, however, the storage modulus exhibits a significant drop, indicating the breakdown of internal structure due to the micro-agitation effect and the reversibility of magnetic clusters.

Based on the above statements, the stiffness of the responsive mixture can be adjusted by applying an external magnetic field. The adjustment range of the storage modulus can be summarized in the gray area in Fig. 11. If a higher structural build-up is desired, the cementitious paste can be exposed to a constant external magnetic field. By adjusting the intensity of the magnetic field, higher stiffness of the cementitious

paste can be obtained. By contrast, if we would like to decrease the stiffness of the paste, a pulsed magnetic field can be applied to the mixture, and then the structural evolution could be slowed down. Therefore, every value in the gray area in Fig. 11 could be possibly achieved for the same cementitious paste containing magnetic particles by using an external magnetic field. As a comparison, the storage modulus of the reference cement paste can only develop according to the black curve in Fig. 11. The active stiffening control can make cementitious materials satisfy different property requirements, contributing to a more reliable and smarter casting process [153]. It can also be used to prevent the leakage of cement grout between formwork joints [154], and even have a potential application in 3D concrete printing [139].

## 6. Conclusions

The origins, evaluation methods, influencing factors and applications of thixotropic structural build-up of cementitious materials are reviewed in this paper. Based on the discussions, the following conclusions can be reached:

- (1) The origin of thixotropic structural build-up of fresh cementitious materials is a physicochemical process with a combined result of colloidal interactions and chemical hydration. After the first contact of cementitious particles with water, the structural evolution of cementitious materials includes three stages, i.e., colloidal network percolation, rigid percolation and rigidification.
- (2) The evolutions of static yield stress and storage modulus (from SAOS test) are useful to evaluate the structural build-up of fresh cementitious materials. The static yield stress is caused by colloidal connections and interparticle bridging by hydration products, whereas the storage modulus refers to the strength of bridges (e.g., C-S-H and/or ettringite) between cement particles. Combining the static yield stress and the SAOS test will be beneficial to monitor the structural build-up of cementitious materials more accurately. Penetration tests can be used to roughly assess the structural build-up of fresh stiff cementitious materials.
- (3) The effect of constituents and mixture proportions on the structural build-up can be quantitatively explained using Eq. (14). Therefore, increasing the particle volume fraction and decreasing the surface coverage and the maximum packing fraction show an increase in the structural build-up of fresh cementitious materials.
- (4) The most significant factor influencing the structural build-up of fresh cement-based materials is w/b. The addition of slag, silica fume and metakaolin has an acceleration effect on the structural build-up, whereas the incorporation of fly ash and limestone powder might slow down the evolution of structural build-up. The addition of superplasticizer decreases the thixotropic structural build-up, with the efficiency depending on the type, structure, dosage, w/b, cementitious materials, as well as the adsorption and the degree of surface coverage. Both in- and organic viscosity modifying agents have a positive effect on the structural build-up. The structuration rate of fresh concrete can be predicted from the structural build-up of the corresponding cement paste, as shown in Eq. (15).
- (5) Considering the non-linear evolution of structural build-up, the lateral pressure of cementitious materials exerting to the formwork can be estimated by Eq. (19). The critical interval time calculated by Eq. (21) can be used as an indicator to guide the multi-layer casting process. The structural build-up of cementitious materials is a potential solution to ensure the sufficient fluidity during extrusion and the high microstructural strength evolution after extrusion. The estimated critical failure time (Eq.

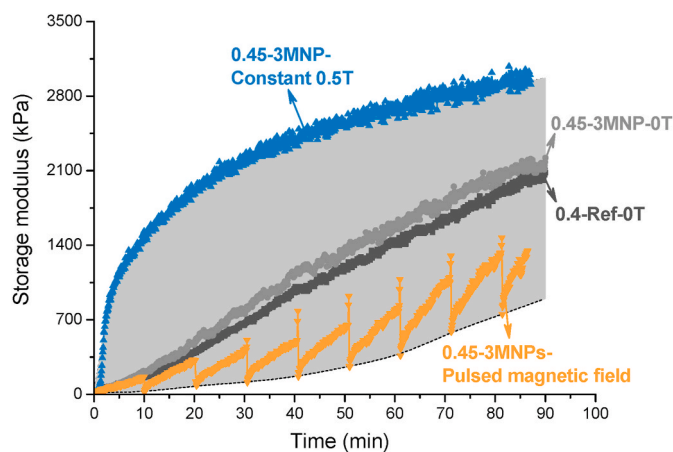


Fig. 11. Representative example of active stiffening control of cementitious paste with w/c of 0.45 and 3% nano-Fe<sub>3</sub>O<sub>4</sub> (200 nm) (adapted from Ref. [153]).

(23)) and maximum interval time (Eq. (25)) have a potential to predict 3D concrete printing.

- (6) Thixotropic structural build-up can have detrimental influence on the simulation of fresh concrete, such as formwork pressure decay, pumping re-initiation, etc. Despite the importance, thixotropy was only rarely considered in simulations, possibly due to the required additional implementation efforts and/or complicated model calibrations. Yet, thixotropic structural build-up shows a huge potential with regard to future numerical investigations.
- (7) The structural build-up of cementitious paste with magnetizable particles can be controlled on-demand by using magnetic field. This active stiffening control can make the cementitious material satisfy different property requirements, contributing to a smarter casting process, minimizing the leakage of cement grout between formwork joints, and even having a potential application in 3D concrete printing.

Since the benefits of thixotropic structural build-up in predicting the stability and formwork pressure during casting, as well as the buildability in 3D printing, the research and knowledge on this area becomes more in-depth and comprehensive in recent years. There is no doubt that on-demand control of structural build-up has a potential to become the mainstream in future studies. However, several challenges remain. For example, advanced techniques and powerful computational models should be utilized to characterize the cohesion forces between particles to interpret the origins of thixotropic structural build-up of cementitious materials qualitatively and quantitatively. The relationships between structural build-up, interparticle interactions and chemical hydration of poly-disperse cementitious suspensions, and the underlying mechanisms should be further investigated, with an emphasis on the suspensions containing nanoparticles and superplasticizer. Regarding the control signals besides magnetic field, the influence and mechanisms of other trigger signals such as temperature and pressure on the thixotropic structural build-up of fresh cementitious materials are required to be clarified. More attention should also be paid to the on-demand control of structural build-up in practical applications of cement-based materials.

#### Declaration of competing interest

The authors declared that we have no conflicts of interest to this work. We declare that we do not have any commercial or associative interest that represents a conflict of interest in connection with the work submitted.

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