A review of concrete properties at cryogenic temperatures: Towards direct LNG containment

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Abstract

This paper provides a "state-of-the-art" review of the pertinent properties of concrete at temperatures lower than -165°C that make it amenable for direct containment of liquefied natural gas (LNG). In addition, the paper presents a brief historical and economic perspective on cryogenic concrete. The permeability, coefficient of thermal expansion (CTE), tensile strain capacity and bond strength to reinforcement are discussed in light of key factors controlling them, including moisture content, aggregate type, etc. Moreover, the effects of cryogenic freeze-thaw cycles on thermal deformation of concrete are highlighted. Generally, the permeability and the CTE are lower while the tensile strain capacity and bond strength to reinforcement are greater for concrete at cryogenic temperatures versus concrete at ambient temperatures. It is concluded that more work is necessary to fully understand thermal dilation and damage growth in concrete due to differential CTE of its components, in order to facilitate development of design methodologies that might be employed to mitigate the associated risks in its eventual utilization for direct LNG containment.

This is an author-created version: regkogbara@cantab.net (RB Kogbara). A definitive version was subsequently published at http://dx.doi.org/10.1016/j.conbuildmat.2013.04.025 in *Construction and Building Materials*, Volume 47, Pages 767 – 770 (2013). The final publication is available at www.sciencedirect.com.

Research highlights

- This is a state-of-the-art review of concrete properties at cryogenic temperatures.
- The properties pertinent for direct LNG containment are identified and discussed.
- The moisture content generally determines the extent of variation of the properties.
- The effects of cryogenic freeze-thaw cycles on concrete deformation are highlighted.
- Some ways to improve concrete durability for cryogenic storage of LNG are suggested.

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Keywords: Cracking; liquefied natural gas; permeability; tensile strain; thermal dilation.

1. **Introduction**

Concrete is the most widely used construction material in the world. Its properties at room, ambient and elevated temperatures are well documented - see, e.g. [1-4]. However, concrete behavior at cryogenic temperatures (i.e. temperatures lower than -165°C) is not fully elucidated. The majority of current liquefied natural gas (LNG) storage tanks, especially in Qatar, utilize 9% nickel steel walls as the primary containment tank due to its greater ductility at cryogenic temperatures compared to normal carbon steel. With the development of the ACI 376-10 code [5] on concrete structures for containment of refrigerated liquefied gases, there may be increasing impetus for tank designs utilizing concrete for primary containment of LNG, which would substantially reduce construction costs. Therefore, a fundamental understanding of the behavior of concrete as it is cooled to cryogenic temperatures is necessary to enhance safety and long-term durability of an 'all-concrete' LNG storage tank. These efforts to understand cryogenic concrete would focus on the mechanism governing thermal dilation and damage growth due to differential coefficient of thermal expansion (CTE) of its components.

Previous research on the properties of concrete at cryogenic temperatures summarized by Krstulovic-Opara [6], which is directly cited in the ACI 376-10 code, show that concrete properties at cryogenic temperatures differ from known behavior at room temperature. The aforementioned work provided a thorough review of the effect of cryogenic temperatures on mechanical and thermal properties of concrete. The work also provided details on the mechanism of water freezing and frost damage, the effect of pore sizes on freezing, and the interaction between freezing water and concrete. The Wiedemann pore model [7], which explains the impact of the above factors on the overall effect of water freezing, and identifies the behavior of freezing water in nine distinct temperature ranges, was also elucidated. However, the effect of bond strength to reinforcement and thermal cycling was not dealt with as the work focused on concrete exposed to a single freezing cycle. Therefore, this paper builds on the work of Krstulovic-Opara [6] and fills in the gaps in the literature as well as documenting newer developments in the field since the last review was published. Beginning with a brief historical and economic perspective on concrete tanks, and a background on ice formation in concrete, this paper focuses on the pertinent properties influencing the design of cryogenic concrete tanks and their controlling factors. The effects of thermal cycling on the pertinent concrete properties are also discussed.

2. **Brief historical and economic perspective on cryogenic concrete tanks**

It has been reported that by 2004, nine tanks using post-tensioned reinforced concrete for the primary containment of LNG have been constructed. The oldest of these tanks has been in continuous service since 1969 [8]. The same tank system were also used for successful storage of liquid oxygen as it proved impervious to liquid oxygen with no metallic liner employed [9]. Figure 1 shows an illustration of a cryogenic tank made up of concrete only, with no metallic liner. Such tanks mainly have a monolithic joint between the primary container walls and slab, a secondary container made with concrete with a moisture vapor barrier applied, and a secondary bottom ideally constructed from a non-metallic material such as Mylar™ sandwich – a poly(ethylene terephthalate) material with very low permeability suited for cryogenic temperatures [10]. A base insulation formed of weatherproofed blocks is also a major feature of such tanks. The expanded perlite insulation between the inner and outer tank wall (Figure 1) acts as a barrier to avoid the possibility of a single containment tank that would be exposed to a strong thermal gradient due to cryogenic temperature on one side and ambient temperature on the other. The secondary container and foundation settings are similar to the conventional 9% nickel steel tanks [8]. However, after expansion of the LNG industry slowed down in the 1980s, marketing efforts decreased leading to a halt in the production of concrete LNG tanks by Cryocrete Inc (then known as Preload) [8].

(Insert Figure 1 here)

Construction of concrete-only tanks was found to be significantly cheaper than 9% nickel steel tanks using cost comparison from three countries as shown in Figure 2. In fact, it has been estimated that while the cost of conventional 9% nickel steel LNG storage tanks in 1999 was about US$400/m^3, concrete-only storage tanks could cost about US$216/m^3 [11]. Thus, there could be a substantial increase in cost savings when the concept of 'all-concrete' tanks becomes established in the LNG market. Further, the ability to construct the primary and secondary containers in parallel using the slip forming technique shortens the construction schedule to around 24 months, compared to 33 – 36 months typical for 9% nickel steel tanks. Such construction techniques also lower operating costs at construction sites [8, 11].

(Insert Figure 2 here)

3. **Concrete properties pertinent for direct LNG containment**

Many properties of concrete improve at cryogenic temperatures [12]. Nevertheless, in the design of concrete for use in direct containment of LNG, certain performance criteria are of utmost importance. One such criterion is the permeability of concrete as it controls the rate by which LNG is lost from the primary container. The permeability of concrete to LNG at cryogenic temperatures is likely dominated by the presence of microcracks since most of the inherent porosity in concrete at cryogenic temperatures is blocked by ice formation [13]. Such microcracks may be caused by interfacial stresses developed within the material due to differences in the CTE of the various components of concrete at cryogenic temperatures –cement paste, aggregates, pore fluids, ice, etc. Furthermore, it is documented that concrete CTE decreases with cooling [6]; however the extent of the decrease is a function of several factors including moisture content, aggregate type and aggregate content of the concrete and the pore size distribution. Moreover, to minimize cracking and limit crack widths, it is beneficial that the concrete has a low CTE and a high tensile strain capacity as well as high bond strength between the concrete and its steel reinforcement. This is because a low CTE would result in smaller strains when there is a temperature gradient, while a high tensile strain capacity would provide the ability to sustain such thermal strains [14].

Historical evidence suggests that the primary LNG containment tank never undergoes a full freezing-and-thawing cryogenic cycle [6] as LNG storage tank systems are in continuous operation such that the tank is never emptied of its contents once filled with LNG. Nevertheless, when some of the LNG is drawn out of the tank, like in the case of a tank at a receiving terminal that is being continually filled and emptied, sections of the tank that are no longer in contact with

LNG are likely to experience increase in temperature [14]. Refilling the tank with LNG again causes rapid temperature decrease of the afore-mentioned parts of the tank. The temperature gradients involved in the above may lead to deleterious tensile stresses. Thus, it is important to consider the effect of freezing-and-thawing cryogenic cycles on concrete even though an inner LNG tank may never undergo a full freezing-and-thawing cycle.

Therefore, from the foregoing, it makes sense that the basic performance requirements of an all-concrete tank for LNG containment are its strength and serviceability requirements. The latter involves cracking and spalling, deformations, vibrations, leakage, and deterioration of concrete reinforcement [15]. Overall, the primary and secondary tanks should be able to maintain structural integrity and liquid tightness, and for the secondary tank, liquid containment. The following sections deals with the aforementioned pertinent properties of concrete and their effect on its use for direct containment of LNG.

3.1. ***Background information on ice formation and frost damage***

The temperature and moisture content play a significant role in influencing the serviceability and durability of concrete for cryogenic applications. The formation of ice in the large capillaries and finer pores in the cement paste, and the subsequent contraction of the ice as temperature falls below freezing point, largely determines changes in concrete properties at cryogenic temperatures [14, 16]. As mentioned earlier, the effects of concrete pore sizes on freezing and the interaction between freezing water and concrete has been extensively discussed together with the Wiedemann's pore model [6].

Noteworthy is the fact that the thermal coefficient of ice is higher than that of the other components of concrete; hence, thermal contraction of ice as temperature is decreased and the capillary effect due to ice formation in the pores are the two main factors that determine the interaction between freezing water and concrete [17]. Moreover, below 0°C, the free energy of gel water (one form in which water exists in hydrated cement paste, the other two being chemically bound water and capillary water) becomes higher than ice enabling the movement of gel water into the capillaries containing ice. Freezing of gel water in the capillaries leads to growth of the ice body in the capillaries. Growth of the ice body with further decrease in temperature, and the increase of surface forces due to the existence of absorbed water layer between the ice surface and capillary wall results in increased matrix stresses and possibly microcracking, which in turn causes considerable expansion of concrete. However, this ice-induced microcracking ceases at about -90°C as water in the nanometric gel pores begin to freeze in the temperature range, -60°C to -90°C, thus slowing down and eventually stopping the growth of the ice body in the capillaries [6].

Overall, it is thought that mechanical properties of concrete improve due to the freezing of water in concrete as ice fills and seals a fraction of pores and microcracks. This is supported by the fact that oven dried concrete exhibits only marginal variations in mechanical properties during cooling as opposed to saturated concrete, whose properties show tremendous increase when cooled to cryogenic temperatures. For instance, the compressive strength of concrete increases threefold when cooled to -170°C. While properties like compressive strength may be greater upon cooling to cryogenic temperatures, such properties are often degraded upon re-warming to room temperature. The expansion of ice leads to microcracking damage which does not manifest until ice melts again. Such damage then manifests as a degradation of strength and stiffness [18].

3.2. ***Permeability of concrete***

The permeability of concrete is the first factor to consider when assessing its suitability for primary containment of LNG. Nevertheless, very little is documented on this property at cryogenic temperatures even in relatively recent reviews on the subject [6, 18]. Permeability tests conducted in the late 1970s and 1980s at a time when there was considerable interest in using concrete for primary containment without the need for a liner indicated that typical concrete mixture designs yield an intrinsic permeability of 10^{-18} m^2 to10^{-19} m^2. The tests were carried out on concrete specimens in contact with pressurized cryogenic liquid [8, 19].

Jackson et al. [8] noted that for permeability considerations, cryogenic concrete mixture design does not require special consideration although aggregates having similar CTE to that of cement paste are preferable. They also recommended water to cement ratios (*w/c*) less than 0.45 and the inclusion of admixtures like silica fume that reduce permeability. The ACI code 376-10 [5] supports the use of such admixtures including fly ash and slag-cement for reducing permeability and increasing durability of concrete as their use can reduce heat development in large sections during hydration and hence, reduce the risk of thermal cracking during construction. Bamforth [14] identified four main factors that affect permeability, albeit based on available literature for concrete at room temperature. These are constituent materials and mixture proportions, degree of compaction, curing regime and extent of cement hydration and moisture content.

On the other hand, the work of Hanaor and Sullivan [13] showed that there are fundamental differences in the mechanisms governing the permeability of concrete at ambient and room temperatures. At low temperatures, filling of the pores with ice, which were hitherto filled with water, impedes the travel of liquid through the pore system, hence reducing permeability [20]. Hanaor and Sullivan [19] corroborate this position as their results indicate the significance of moisture content among factors affecting permeability as the formation of ice serves as a partial barrier to fluid flow by reducing the effective porosity. This work showed that aggregate type played a dominant role amongst factors affecting permeability at cryogenic temperatures, which also included air-entrainment and *w/c* in addition to moisture content mentioned above. The importance of aggregate type suggests that the mechanism involved in concrete permeability at cryogenic temperatures may be associated with the thermal incompatibilities of the aggregate and the cement paste. Such incompatibility may lead to microcracks, which provide enhanced flow paths for the permeating fluid thus increasing permeability. Thermal stresses in concrete are a function of the differential thermal strains, which depend on the CTE differential and the differences in elastic moduli. Hence, a judicious choice of aggregates, for example rounded lightweight aggregate is essential for cryogenic concrete to achieve target permeability coefficients of 10^{-18} - 10^{-20} m^2. The use of lightweight aggregate is suggested principally because of the enhanced bond between the coarse aggregate and the matrix which is attributed to elastic stiffness compatibility [5, 19]. It stands to reason, therefore, that it is due to the aforementioned findings that the relevant ACI code recommends that concrete aggregates for use in primary concrete containment structures should have a low coefficient of thermal expansion but not so low that incompatibility with the cement matrix can lead to cracking at the aggregate/matrix interface and consequent increased permeability [5].

Bamforth [14] also indicated that lightweight concrete exhibited appreciably lower water and gas permeability coefficients compared to normal weight concrete. It was observed that of different mixtures tested, lightweight concrete with sintered PFA aggregate was the most suitable for cryogenic applications as it exhibited the lowest permeability coefficient at ambient temperature (Table 1). It also showed a combination of high strain capacity, low Young's modulus of elasticity and low CTE. The data in Table 1 also show the difference between water and gas permeability coefficients for concrete samples. The difference between water and gas permeability coefficients was larger at lower permeability values than at higher permeability values. This supports the position of Klinkenberg [21] that the difference between liquid and gas permeability coefficients were considerable for media of low permeability.

Table 1. Water and gas permeability coefficients for different concrete mixtures at ambient temperature [14]

Permeability coefficients and mixture details	Sample Number	AEC Air entrained control mixture	HS High strength mixture	LW Lightweight mixture	PFA Pulverised fuel ash mixture
Gas permeability coefficient (k_g) obtained at a pressure of 1.03 N/mm^2 (x10^{-18} m^2) (the mean in the next column is geometric mean based on two values for each sample)	1	11.44 / 9.81	5.61 / 3.09	0.39 / 0.50	2.80 / 5.97
	2	18.31 / 12.49	0.77 / 0.29	0.43 / 0.20	2.99 / 3.57
	3	14.46 / 15.26	0.95 / 0.31	0.45 / 0.79	2.11 / 1.96
	Mean	(13.35)	(1.02)	(0.43)	(3.01)
Water permeability coefficient (k_w) obtained at a pressure of 1.03 N/mm^2 (x10^{-18} m^2)	1	3.74	5.44 x 10^{-2}	3.27 x 10^{-2}	5.07 x 10^{-2}
	2	0.81	3.16 x 10^{-2}	2.59 x 10^{-2}	2.71 x 10^{-2}
	3	4.52	2.68 x 10^{-2}	46.05 x 10^{-2}	4.09 x 10^{-2}
	Mean	(2.39)	(3.58 x 10^{-2})	(7.31 x 10^{-3})	(3.83 x 10^{-2})
Water permeability coefficient (m/s) (same as k_w above but in units of m/s)	1	3.67 x 10^{-11}	5.34 x 10^{-13}	3.21 x 10^{-14}	4.97 x 10^{-13}
	2	7.80 x 10^{-11}	3.07 x 10^{-13}	2.54 x 10^{-14}	2.66 x 10^{-13}
	3	4.43 x 10^{-11}	2.63 x 10^{-13}	45.15 x 10^{-14}	4.01 x 10^{-13}
Ratio of gas to water permeability (k_g / k_w)	-	5.6	28.5	58.8	78.5
Ordinary Portland Cement	-	440	475	505	335
Pulverised Fuel Ash	-	-	-	-	115
Gravel (20 – 5 mm)	-	1,100	1,160	-	1,100
Lytag (12 mm) (sintered PFA lightweight aggregate)	-	-	-	780*	-
Sand	-	600	625	495	565
Water	-	170	155	205**	185
Melment L10 (superplasticiser)	-	-	7 litres	-	-
Sika AEA (air entraining admixture)	-	0.31 litres	-	0.18 litres	-
Water/cement ratio	-	0.39	0.33	0.41	0.41
Slump (mm)	-	75	80	70	70
Air content (%)	-	4.8	1.3	6.4	0.8
28-day Compressive Cube Strength (MPa)	-	51.9	90.3	49.5	57

*Saturated surface dry value with 12% moisture content **Free water content, excluding water absorbed into the aggregate

There is a paucity of data on liquid permeability of concrete at cryogenic temperatures. However, limited data from Bamforth [14] on gas permeability over a range of temperatures, which incorporated some data from Eaking et al. [22] for comparison, showed that reduction in temperature to as low as -165°C has a relatively small effect on the specific gas permeability, generally less than an order of magnitude (Figure 3). This small effect on permeability is probably due to the small decrease in the volume of overall void space caused by increase in volume of freezing water, especially in unsaturated concrete. The decrease in void space is quite small with no significant reduction in the voids that constitute flow paths for the gas. Figure 3 also shows that while the specific gas permeability clearly reduces with decreasing temperature, the effect of temperature decrease is small compared to the effects of moisture content, mixture proportions and curing conditions. The lightweight aggregate mixture still showed lower permeability coefficient compared to the other mixtures with the same curing condition (Figure 3), which corroborates the position of Hanaor and Sullivan [19].

(Insert Figure 3 here)

It can be deduced from the foregoing that the basic methods of creating relatively impermeable concrete for cryogenic applications are the use of lightweight aggregates, air-entrained admixtures, pozzolans such as fly ash and reduction in *w/c* ratio. The effect of air-entraining admixtures in concrete is to stabilize air bubbles by reducing the surface tension at the air-water interface. As water in the pores of concrete freezes, it expands, creating pressure on the remaining liquid, which creates stress on the concrete, and in turn causes cracks. Entrained air voids provide extra space for the water to flow into (and freeze) so that it does not generate large expansive stresses that cause damage in the concrete. Pozzolans densify, strengthen, and increase the insoluble fraction of the microstructure. A low *w/c* reduces the magnitude and connectivity of the pore network, thereby limiting the permeability and the influence of pore ice [16].

3.3. ***Coefficient of thermal expansion (CTE)***

The CTE of concrete decreases with cooling, with the response depending largely on the relative humidity at which the concrete is stored [6]. In general, a critical moisture content corresponding to storage at a relative humidity of about 86% has been identified. The response of oven dry concrete and concrete stored at a relative humidity less than 86% is governed by the type of aggregate as shown in Figure 4 [23]. While the behavior of water-saturated concrete and concrete with relative humidity greater than 86% is governed by the moisture content. There is a sudden decrease in the CTE between 0ºC to -75ºC depending on the moisture content. Overall, concrete with higher moisture content expands with cooling below 0ºC, while concrete with medium moisture content contracts with cooling [6, 24].

(Insert Figure 4 here)

It is important to note that as concrete (especially non air-entrained concrete) is cooled below 0ºC it may exhibit a gradual contraction, except in the temperature range, -20ºC to -70ºC, where it expands before resuming its contraction. This is shown in Figure 5a, extracted from Marshall [16]. The moisture content of the concrete largely influences the amount of expansion and temperature change: dry concrete does not expand, but wet concrete does [16]. This is attributed to gradual filling of the finer pores with ice, which were hitherto filled with super cooled water, and the attendant internal pressure increase within the aforementioned temperature range. Hence, towards -70ºC when the finest pores become frozen, the concrete begins contracting again [18]. Also noteworthy is the observation that the wet concrete in Figure 5a contracts more than the dry concrete at the beginning of cooling in contrast to the trend shown in other works, in which the contraction is almost the same or slightly higher in dry concrete [6, 25, 26]. The deviation from the usual trend is probably due to differences in the cement type compared, as well as the type of aggregate used. The slightly higher contraction at the beginning of cooling of water-saturated concrete made with blast furnace slag-cement compared to normally stored concrete made with Portland cement, shown in Figure 5b [27] further illustrates the above position. Furthermore, the influence of moisture content on CTE is corroborated by experimental results of Rostasy et al. [28], which showed the influence of *w/c* and the pore water content on the expansion exhibited by wet concrete (Figure 6). The data demonstrates that increasing the *w/c* increased the extent of the expansion phase, as did increasing the test moisture content.

(Insert Figures 5 and 6 here)

It is well known that at room temperature, the CTE of concrete depends on the composition of the mixture and on its moisture state at the time of the temperature change. The effect of mixture composition is due to differences in the CTE of hydrated cement paste and aggregate, since the CTE of concrete is the resultant of the two values. The linear CTE of hydrated cement paste varies between 11 x 10^{-6} and 20 x 10^{-6} per ºC [29] (depending primarily on the moisture content [30]) and is higher than the CTE of aggregates (see Table 2). This difference in CTE may have a deleterious effect during large temperature swings. Moreover, at cryogenic temperatures, concrete is a complex composite consisting of aggregates (with pores), cement paste (with pore radii ranging from nm – mm and several distinct solid phases), pore liquid, pore vapor, interfaces, dissolved species in the pore fluid, and ice. Hence, differential CTE of the various phases present in concrete may lead to the development of interfacial stresses, which may in turn cause microcracking. However, there is inadequate information on this subject at cryogenic temperatures in the literature. Thermal strains are by far the most significant of the strains induced in the wall or base of an LNG concrete tank as cooling from ambient temperature to -165ºC can result in contraction of 1500 microstrain compared to about 300 microstrain due to a prestress of 10 N/mm^2 from steel [14]. Therefore, an in-depth knowledge of the thermal behavior of concrete at cryogenic temperatures is necessary for the design of concrete tanks with predictable performance in order to mitigate the extent of microcracking due to differential CTE in the material.

Table 2. Typical linear thermal expansion coefficient for common Portland cement concrete components at ambient temperature [31]

	Coefficient of Thermal Expansion	
	10^{-6}/°C	**10^{-6}/°F**
Aggregate		
Granite	7 – 9	4 - 5
Basalt	6 – 8	3.3 - 4.4
Limestone	6	3.3
Dolomite	7 – 10	4 - 5.5
Sandstone	11 – 12	6.1 - 6.7
Quartzite	11 – 13	6.1 - 7.2
Marble	4 – 7	2.2 - 4
Cement Paste (saturated)		
w/c = 0.4	18 – 20	10 - 11
w/c = 0.5	18 – 20	10 - 11
w/c = 0.6	18 – 20	10 - 11
Concrete	7.4 – 13	4.1 - 7.3
Steel	11 – 12	6.1 - 6.7

3.4. ***Tensile strain capacity***

Temperature gradients in concrete leads to tensile stresses and when such stresses exceed the tensile strain capacity, cracks are formed in the concrete. Thus, the tensile strain capacity is the maximum tensile strain that concrete can withstand without a continuous crack forming. It has been reported that the tensile strain capacity ('cracking strain') at ambient temperature is affected by the moisture condition of the concrete. Compared to wet concrete, the cracking strain of oven-dried concrete increases due to increase in the energy required for crack growth. The cracking strain was also found to decrease with increase in the *w/c*. However, there was no significant effect of cement type (types I, III and V) on the cracking strain at ambient temperature [32].

The type of aggregate affects the tensile strain capacity. For example, lightweight concrete has a higher tensile strain capacity than normal weight concrete upon cooling to cryogenic temperatures. Bamforth [14] observed that at ambient temperature, the strain capacity of lightweight concrete was about 50% higher than that of normal weight concrete but it became double at -165ºC (Figure 7). This implies that lightweight concrete can sustain twice the thermal deformations normal weight concrete would before cracking [14, 33]. The higher strain capacity of the lightweight concrete is because the Young's modulus of elasticity of normal weight concrete is roughly twice that of lightweight concrete. Thus, prestressing of lightweight concrete results in proportionately greater resistance to thermally induced cracking than normal weight concrete [33]. Similarly, it is important to point out that reinforcement adds tensile strain capacity to the concrete and helps control crack width due to temperature gradients.

(Insert Figure 7 here)

3.5 ***Bond strength to reinforcement***

The bond strength to reinforcement of concrete is related to the tensile strength. Hence, it is influenced by the same factors that affect the tensile strength, such as moisture content and aggregate type. Moreover, the design of reinforcement for crack control depends on knowledge of the tensile behavior of the reinforcing steel and concrete and the bond strength that can be developed between the two materials. As with the other properties discussed in preceding sections, the bond strength to reinforcement increases with decreasing temperature and even doubles the ambient temperature value at -160ºC [34, 35]. The bond strength of plain bars in water-saturated concrete even exhibits a higher increase at -196ºC. The ratio of the bond strength at the aforementioned cryogenic temperature to that at room temperature was reported to be about 6 for air-entrained concrete and 2 – 3 for non-air entrained concrete [35]. The results from the aforementioned studies [34, 35] have been summarized by Bamforth [14], as shown in Figure 8. The yield and tensile strength of reinforcing steel also increases as temperatures drop to cryogenic values [12, 36]. Overall, at low temperatures, moisture content plays the most relevant role on the bond strength: air-dry concrete gives the lowest bond resistance, while water-saturated and sealed concrete behave practically in the same way [37].

(Insert Figure 8 here)

In the design of an all-concrete LNG tank, the thermal deformation arising from temperature gradients during cool down and any warming due to reduction in level (withdrawal) of LNG in the tank is an important factor to be considered. Consequently, knowledge of the thermal deformation behavior of concrete and the reinforcing steel is crucial. Figure 9a extracted from Elices [38] shows that reinforcing steel exhibits an almost linear thermal deformation with temperature while water-saturated concrete shows a completely different behavior. This difference may lead to internal stresses which results in the steel being 'overstressed' while the concrete suffers additional compression. Nevertheless, it can be deduced from Figure 9b that in the presence of compressive stress, especially above 15 MPa, water-saturated concrete exhibits an almost linear thermal deformation. Thus, since prestressing results in compressive stress on concrete, it would be useful in mitigating damage arising from differences in the thermal deformation behavior of steel and concrete, as steel and concrete may behave in almost the same way with prestressed concrete [25].

(Insert Figure 9 here)

4. **Other properties**

It should be noted that several properties of concrete are considered in the design of concrete for cryogenic storage of LNG. This paper focuses on the most pertinent properties especially as previous review efforts on cryogenic concrete have discussed most of the other properties. These other properties include compressive strength, creep, Poisson's ratio, Young's modulus of elasticity, thermal conductivity, and specific heat. Information on these properties has been extensively reviewed and can be found in two of the most recent reviews on cryogenic concrete – Krstulovic-Opara [6] and Dahmani et al. [18]. In summary, the compressive strength, Poisson's ratio, Young's elastic modulus and thermal conductivity increase with decreasing temperatures while creep and specific heat decreases with decreasing temperature. Just like most of the properties considered in this work, the compressive strength, Young's elastic modulus, thermal conductivity and specific heat are strongly dependent on moisture content. The Young's modulus and the two thermal properties also depend on the aggregate used [16].

5. **The effects of thermal cycling**

Repeated freezing and thawing of moisture contained in the pores, microcracks, and cavities of concrete is a source of damage in concrete. As the concrete freezes, water in the free state in cracks changes into ice, accompanied by volumetric increase, thus expanding the recess in which freezing occurs [18]. However, upon thawing, the contraction is smaller and more rapid in relation to temperature than the original freezing expansion [16]. This leads to a reduction in strength and stiffness below the initial value and the severity increases with the moisture content of the concrete. Further freeze-thaw cycles have a further marked effect [18]. The extent of the resulting damage from thermal cycling depends on the rate of cooling. It has been summed up that the influence of thermal cycling on concrete depends on a number of factors including the quantity of air contained in the concrete, rate of change of the temperature, size and shape of the concrete piece tested, age, porosity, water/cement ratio, moisture content, and method of curing [16].

5.1 ***Effect on permeability***

After initial cool down during filling of LNG tanks, they are maintained at low temperatures, but the tanks or parts of it may be returned to ambient temperature if there is need for inspection and maintenance and during withdrawal of LNG. It has been demonstrated that the effect of thermal cycling on gas permeability of normal weight concrete was far less than that of lightweight concrete. Bamforth [14] observed an increase by a factor of 2 for the former and 100 for the latter after slow (4ºC per hour from 20ºC to -165ºC) thermal cycling. The increase was even greater with rapid temperature change. The large permeability increase of the lightweight mixture is attributed to disruption due to ice formation since the lightweight mixture has higher moisture content than normal weight mixture due to its porous aggregates. The total porosity increases with freeze-thaw cycling and the porosity coarsens, probably due to formation of percolated internal cracks. This pore coarsening leads to increases in permeability since the percolated cracks provide more flow paths for fluid transport [39]. Figure 10 from Wardeh et al. [39] show gas permeability evolution during freeze-thaw cycles in relation to the frost damage factor, which is related to the reduction of Young's elastic modulus with concrete damage. It is clear that the intrinsic permeability increases progressively with repeated freezing and thawing. However, there are indications that prestressed concrete permeability may not be severely affected as limited evidence suggests that confinement of the concrete prevents the damage associated with ice formation [14].

(Insert Figure 10 here)

5.2 *Effect on coefficient of thermal expansion*

Freeze-thaw thermal cycling of water-saturated concrete results in irreversible strains which indicate micro-cracking as a result of frost attack. The irreversible strains and expansion of the concrete are progressively increased with repeated freeze-thaw cycles. Figure 11 shows the evolution of thermal strains during two thermal cycles between 20ºC and -165ºC for unloaded and loaded water-saturated concrete. Loading of the concrete entailed applying a compressive stress of 15 MPa by means of hydraulic jacks. The temperature range, -20ºC to -60ºC, in which water transforms into ice in pores of small diameter is identified as the problematic region where expansion occurs [25, 40]. The expansion is even more pronounced in the said temperature range when the concrete returns to room temperature, leading to concrete deterioration (Figure 11). Comparison of the behavior of water-saturated and dried concrete in Figure 11a indicates that higher moisture content and *w/c* leads to greater expansion of concrete during thermal cycling.

Hence, it was suggested that the removal of weakly bound water in concrete is the way forward for making cryogenic frost-proof concrete [18]. However, with a compressive load of 15 MPa, similar to the compressive load induced by prestressing, water-saturated concrete exhibits highly diminished non-linear effects in the aforementioned temperature range and a compressive irreversible strain increase [25]. This corroborates the findings of Musleh [41] that prestressing of concrete improves its effective durability against cyclic freezing and thawing.

(Insert Figure 11 here)

It should be noted that deterioration of concrete after freeze-thaw cycles is exacerbated by the relatively large difference in the CTE of cement paste and aggregate. Freeze-thaw cycling tests on concrete and concrete constituents demonstrated that the maximum expansion and residual strains after five cycles were larger in the order of cement paste, mortar, concrete and aggregate (Table 3). Hence, it is thought that strain behavior and deterioration of concrete are mainly due to cement paste and interfacial fracture between aggregate and cement paste. Thus, durable cement paste of lower *w/c* or lower moisture content, and air entraining agent are necessary to increase the freeze-thaw durability of concrete [40]. It is well known that air entrainment diminishes the damage to concrete from cryogenic freeze-thaw cycling. Montfore and Lenz [42] did not record any observable effect on air-entrained concrete after 10 freeze-thaw cycles between 24ºC and -157ºC. Air entrained bubbles in concrete buffers changes in volume of water as it changes into ice, and prevents concrete damage. Hence, air-entrained concrete contracts after ice nucleation, which offsets the thermal expansion mismatch with ice, so cracking is reduced, as opposed to non-air entrained concrete, which experiences significant expansion after ice nucleation [43]. Overall, it appears that the absolute value of the CTE of water-saturated concrete reduces with the progress of freeze-thaw cycles, while that of dried concrete and air-entrained concrete remains unchanged over several freeze-thaw cycles [44].

Table 3. Properties of concrete constituents during freeze-thaw cycles [40]

Type of concrete constituent	Coefficient of thermal expansion ($x10^{-6}$/°C)		Maximum expansion ($x10^{-6}$)	Residual strain after the 5th cycle ($x10^{-6}$)
	-10ºC	-45ºC		
Aggregate	4.6	3.0	-	0
Paste	17.5	-122.9	1367	908
Mortar	13.0	-66.9	703	582
Concrete	6.9	-25.7	376	445

5.3 ***Effect on tensile strain capacity***

Moist concrete exhibits larger dilation and residual strains than air-dried concrete after gradual cryogenic freeze-thaw cycles [45]. Great losses of strength (both tensile splitting strength and compressive strength) only occur after thermal cycling if concrete was initially water-saturated [28]. The splitting tensile strength of air-dried concrete cycled through cryogenic temperature does not undergo any significant damage, whereas that of moist concrete may exhibit up to a 20% loss in strength. This sharp drop in splitting tensile strength of moist concrete can be caused by de-bonding observed at the interface between aggregate and cement paste [28]. A similar observation was made with sulfur concrete cycled between room and liquid nitrogen temperatures as fracture surfaces showed clear de-bonding of SiO_2 particles from sulfur after cycled samples showed five times less compressive strength compared to non-cycled samples. The observed de-bonding was attributed to large differences in the CTE of sulfur and aggregate (similar to the remark made in section 5.2) and it initiates at some yet unknown temperature where the induced strain was sufficient for the sulfur to go from elastic to plastic behavior [46].

It has been reported that during freeze-thaw cycles between room temperature and -25°C, the tensile strain for below 0°C temperatures increases progressively with the number of freeze-thaw cycles. At the end of each thermal cycle, irreversible tensile strains develop due to the observation that some concrete constituents have plasticity in tension during freezing and thawing [47]. This plastic tensile strain increases with the number of thermal cycles. Again, air-entrainment was found to lead to much lower freeze-thaw cycle plastic tensile strain as sufficient pores for ice expansion are provided in air-entrained concrete thereby preventing the occurrence of cracks [47].

5.4 *Effect on bond strength to reinforcement*

There is a dearth of literature on the effects of cryogenic freeze-thaw cycles on concrete bond strength to reinforcement. Nevertheless, freeze-thaw cycling between 25°C and -25°C in pull-out tests on the effect of frost on bond properties between concrete and steel reinforcement showed pull-out failures at steel stresses below the yield strength. It was observed that the bond capacity between steel and concrete was reduced by 14% and 50%, corresponding to 25% and 50% decreases in compressive strength from freeze-thaw cycling, respectively [48].

6. **Conclusions**

The key properties of concrete pertinent to its use for direct containment of LNG have been identified and reviewed. These properties include permeability, CTE (of all concrete components), tensile strain capacity, and bond strength to reinforcement. The effects of one-time cooling concrete from ambient to cryogenic temperatures and thermal cycling on these properties were highlighted. From published literature, it is obvious that variation in concrete properties from ambient to cryogenic temperatures largely depends on the moisture content of concrete. The moisture content generally determines the extent of variation of a given property. Generally, the permeability and the CTE decreases, while the tensile strain capacity and bond strength to reinforcement increases over their ambient temperature values when cooled to cryogenic temperatures. The main mechanism governing this is the formation of load bearing ice in concrete pores. Furthermore, the tensile and compressive strength, Poisson's ratio, Young's elastic modulus and thermal conductivity increase with decreasing temperatures. Creep and specific heat both decrease with decreasing temperature.

Based on the cryogenic temperature behavior of the pertinent concrete properties as well as the detrimental effect of cycling between ambient and cryogenic temperature on them, a number of ways to prevent thermal cracking and hence improve concrete durability for use in cryogenic storage of LNG are suggested. These include using (lightweight) aggregates with a high tensile strain capacity and low CTE that is not incompatible with the CTE of cement paste, use of air-entraining admixtures, and pozzolans like fly ash; and using a low *w/c*. Other approaches to prevent thermal cracking are, proper prestressing, which helps to keep the concrete water-tight and free from major cracks; and keeping the concrete continually moist for a reasonable period such that it continues to hydrate and heals any minor cracks present.

The review of literature on the subject matter shows that there is inadequate information on thermal stresses due to CTE mismatch among the different phases present in concrete at cryogenic temperature, which may result in microcracking. Studies are in progress in the authors' laboratories to measure and model thermal dilation of concrete at cryogenic temperatures and to develop a model for predicting potential damage growth in concrete due to CTE mismatch induced stresses. Ultimately, design methodologies that might be employed to reduce the risk or mitigate the extent of microcracking due to differential CTE of components in concrete at cryogenic temperatures would be developed.

Acknowledgements

This publication was made possible by an NPRP award (NPRP No. 4-410-2-156: *Thermal Dilation and Internal Damage of Cryogenic Concrete used for Direct LNG Containment*) from the Qatar National Research Fund (QNRF – a member of The Qatar Foundation). The statements made herein are solely the responsibility of the authors. The authors also thank Ms. Syeda Rahman, Mr. Boback Parsaei, Ms. Noor Masad and Dr. Emad Kassem of Zachry Department of Civil Engineering, Texas A&M University, College Station, USA for their support.

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Figure captions

Figure 1. Cryogenic concrete tank (a) cross section, and (b) details [8]

9% Ni – 9% nickel steel tank, ACLNG – all concrete LNG tank

Figure 2. Cost comparison between all-concrete tanks and 9% Ni steel tanks [8]

AEC – air entrained control mixture, HS – High strength mixture, LW- lightweight mixture, PFA – PFA mixture,

Ref. 29: from Eaking et al. [22]

Figure 3. Gas permeability coefficients over a range of temperatures for concretes subjected to different curing conditions [14]

Figure 4. Change in the coefficient of thermal expansion with temperature for concretes with dry to moderate moisture content [23].

PZ: Portland cement stored at 20°C and 65% RH, HOZ: blast furnace slag cement

Figure 5. Variation of (a) CTE with wet and dry concrete (b) thermal strain with water-saturated and normally stored concrete [16, 27].

PZ - Portland cement, HOZ - blast furnace slag cement, 2.5 K/min and 1 K/min – cooling rates in Kelvin, 1:4:0.8; 1:3:0.5 and 1:6:0.5 - cement: sand: water (i.e. *w/c* of 0.8, 0.5 and 0.5, respectively)

Figure 6. Influence of initial *w/c* and moisture content on thermal contraction of Portland cement and blast furnace cement (a) mortars and (b) concrete [28]

AEC – air entrained control mixture, HS – High strength mixture, LW- lightweight mixture, PFA – PFA mixture,

1 microstrain = 0.0001% strain

Figure 7. Variation of tensile strain capacity with temperature [14]

Ref. 11: Yemane et al. [35]; Ref. 17: Goto and Miura [34]

Figure 8. The influence of temperature on the bond strength of concrete to reinforcement [14]

Figure 9. Thermal behavior of (a) unloaded water-saturated concrete, and (b) loaded water-saturated concrete loaded [38]

$D_f = 1 - E/E_0$; where, E is the Young's elastic modulus after N freezing–thawing cycles and E_0 the initial elastic modulus of the non-damaged (non-cycled) material. The elastic modulus is used as a measure of the damage factor because it decreases significantly once internal cracking resulting from freeze-thaw damage occurs.

Figure 10. Gas permeability as a function of frost damage factor [39]

Figure 11. Thermal strains of (a) unloaded water-saturated and dried concrete, and (b) loaded water-saturated concrete along two thermal cycles [25]

Figure 1

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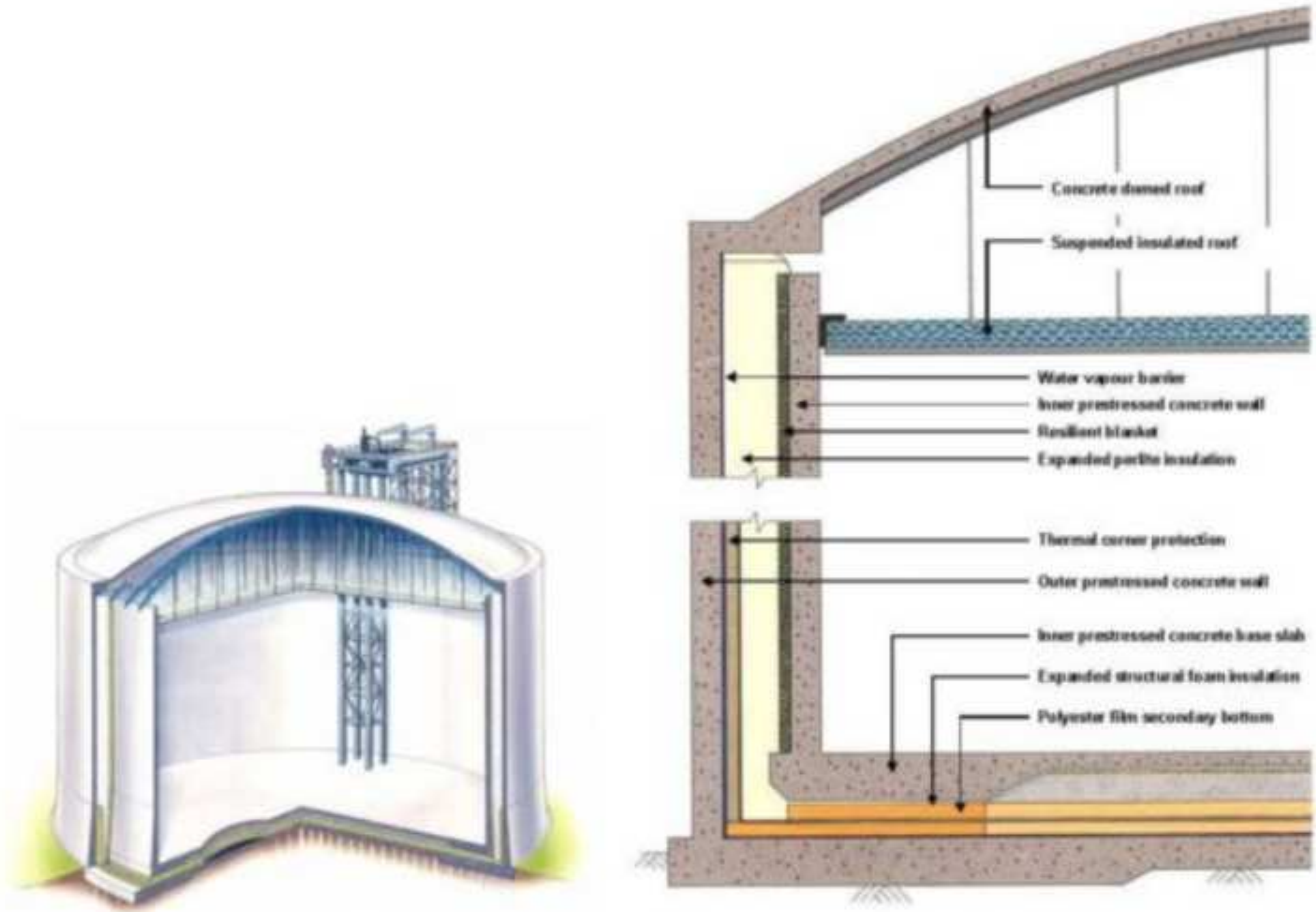

(a) (b)

Figure 2
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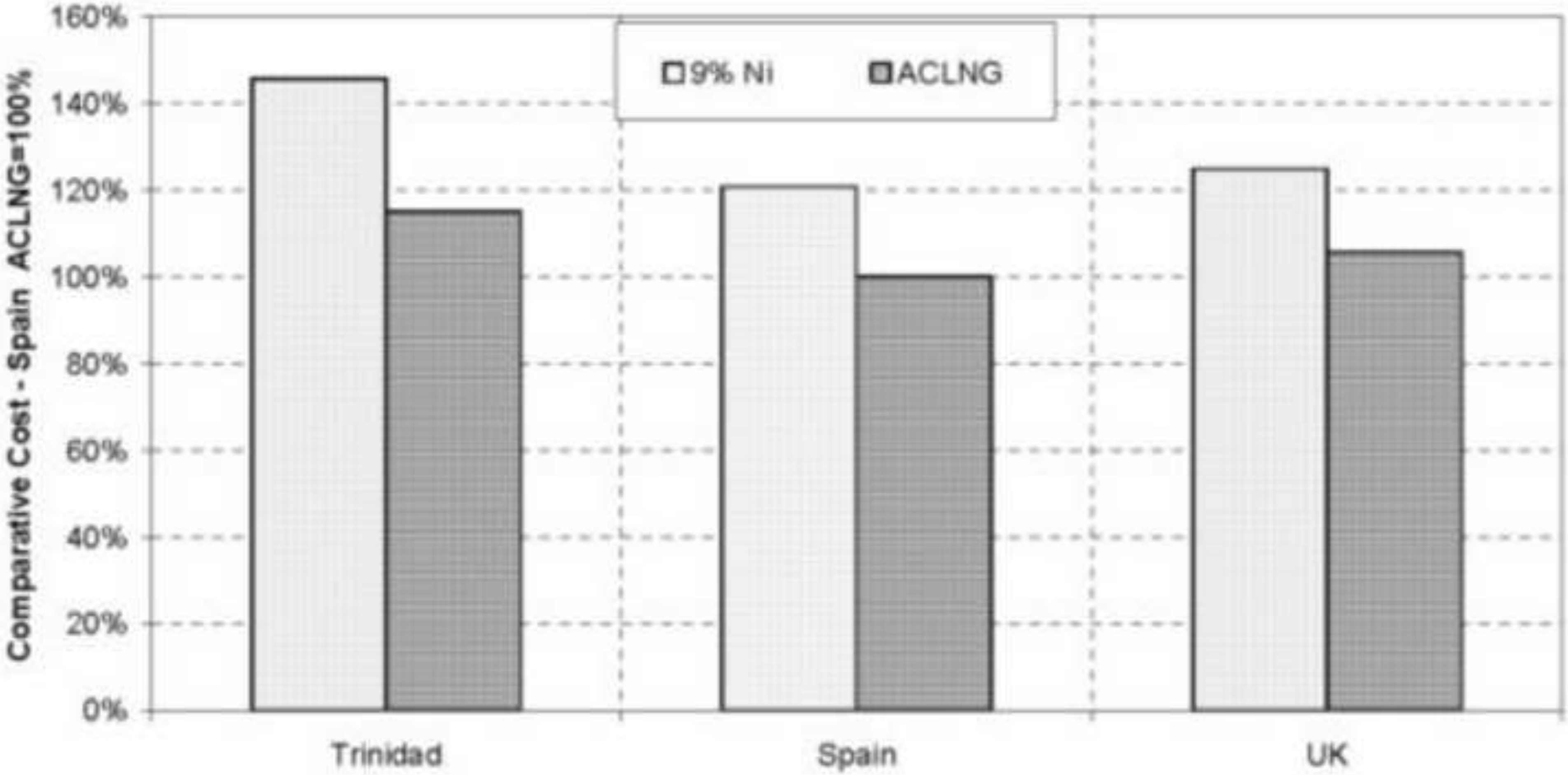

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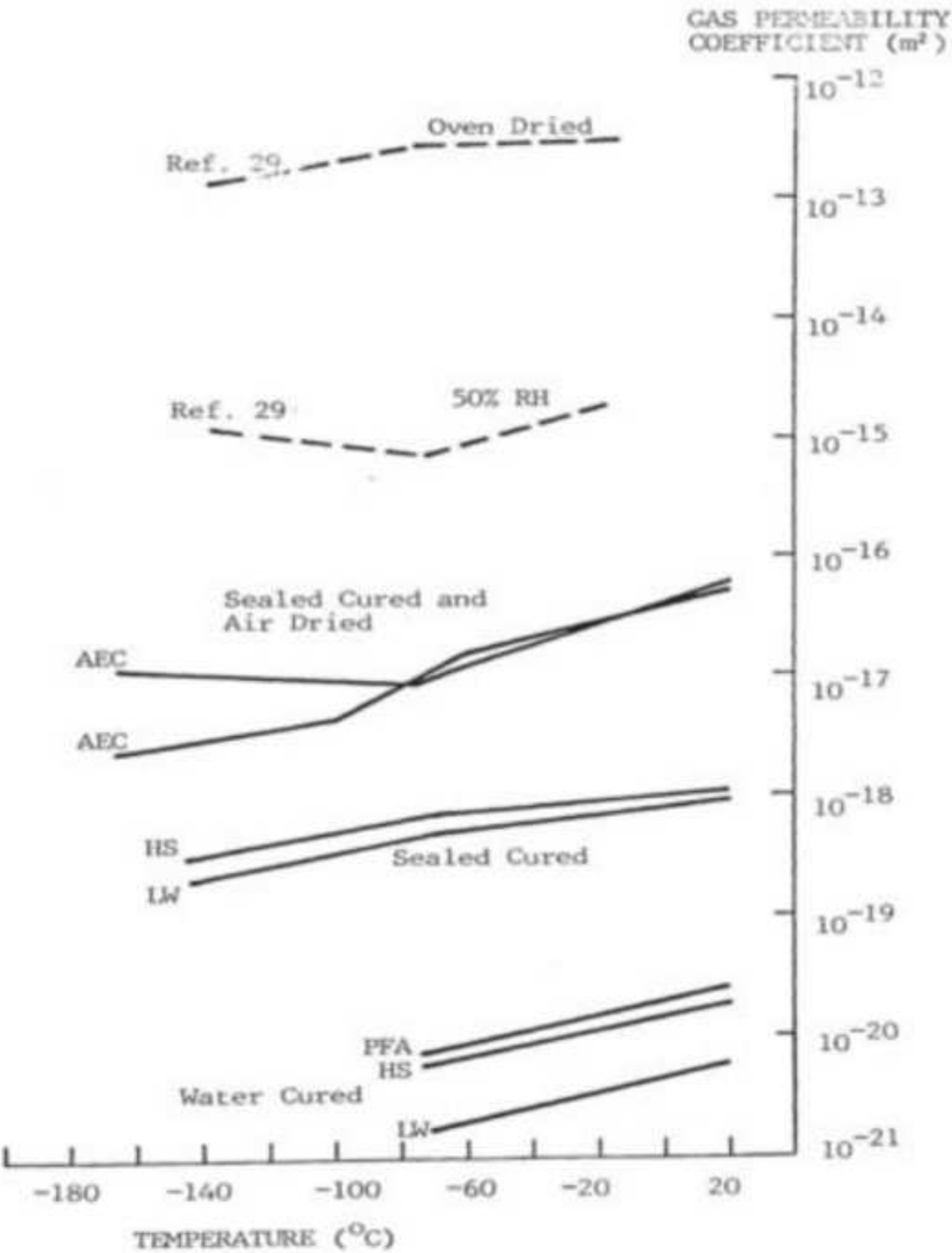

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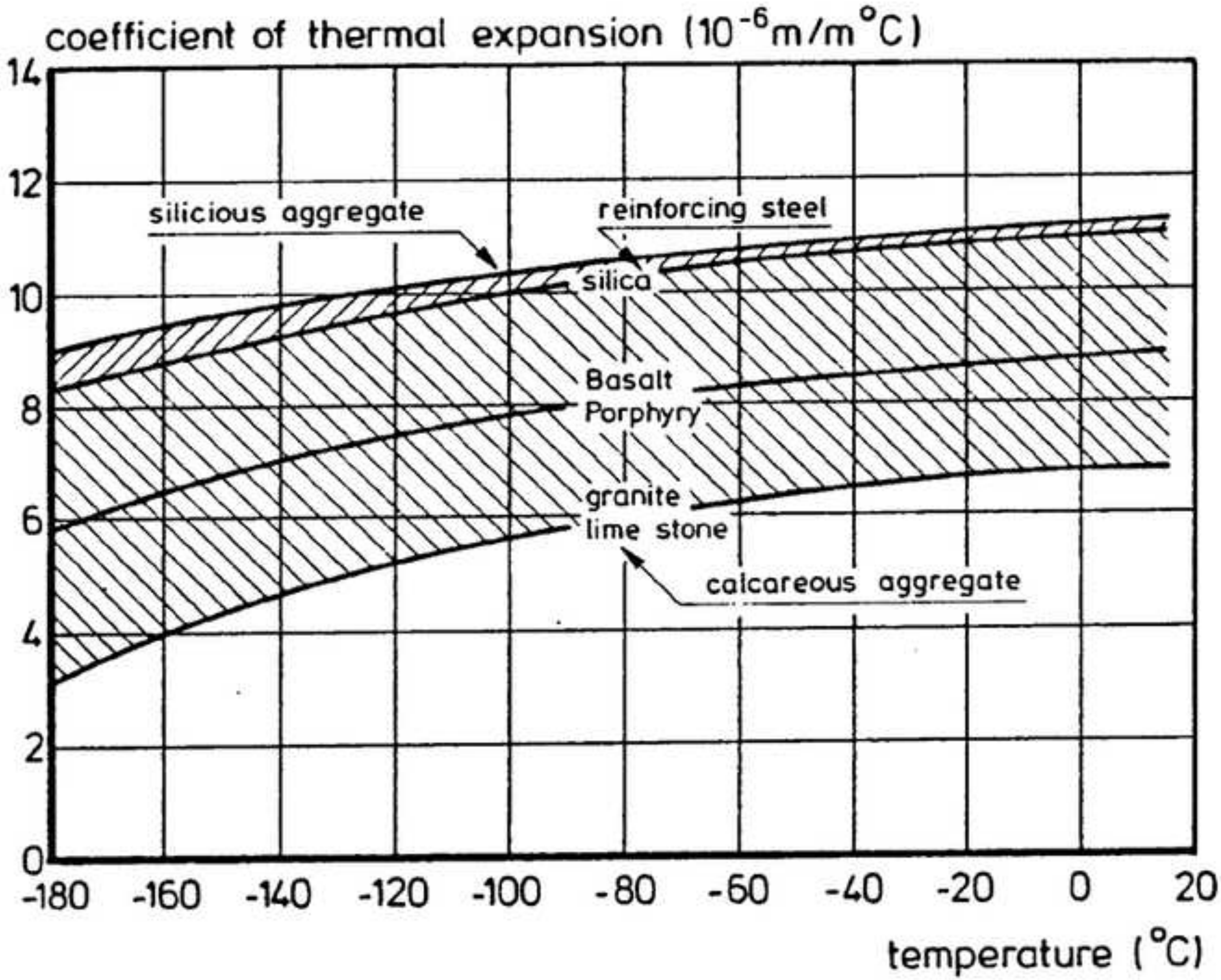

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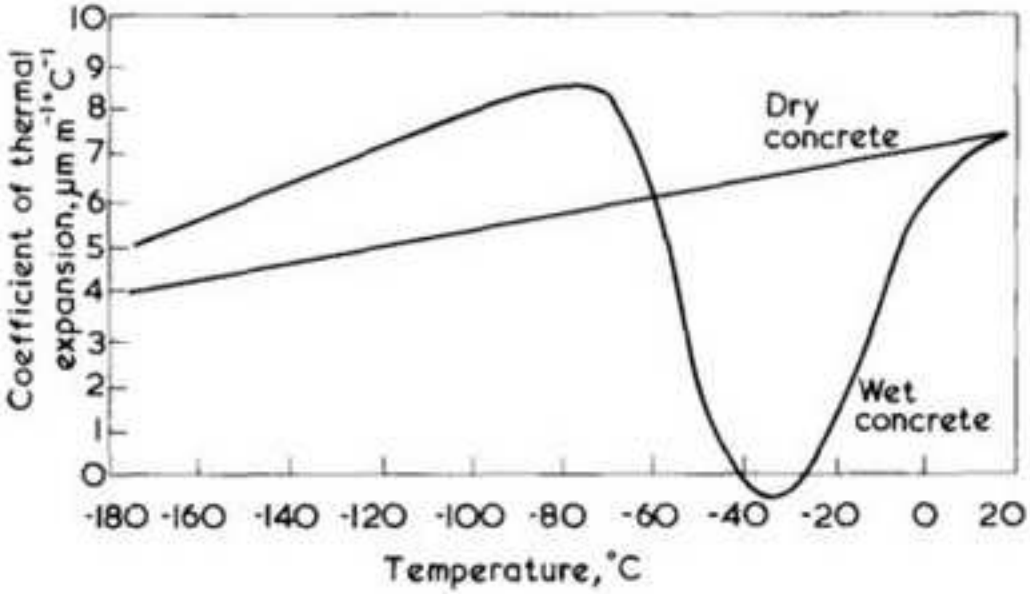

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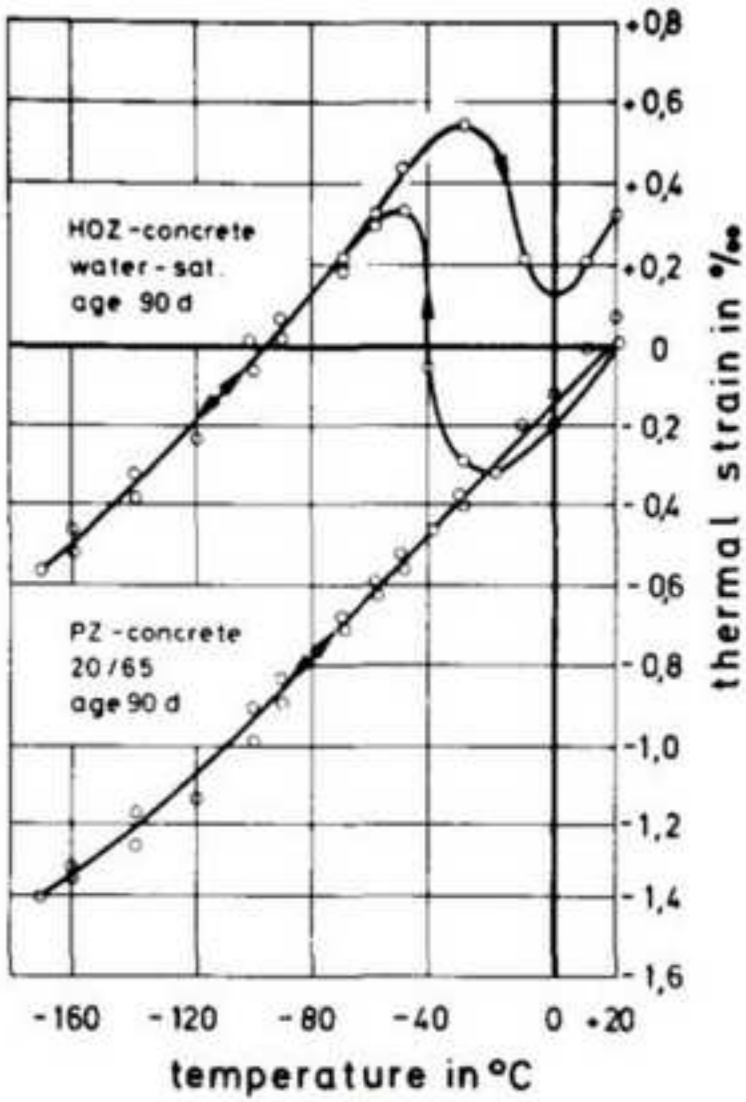

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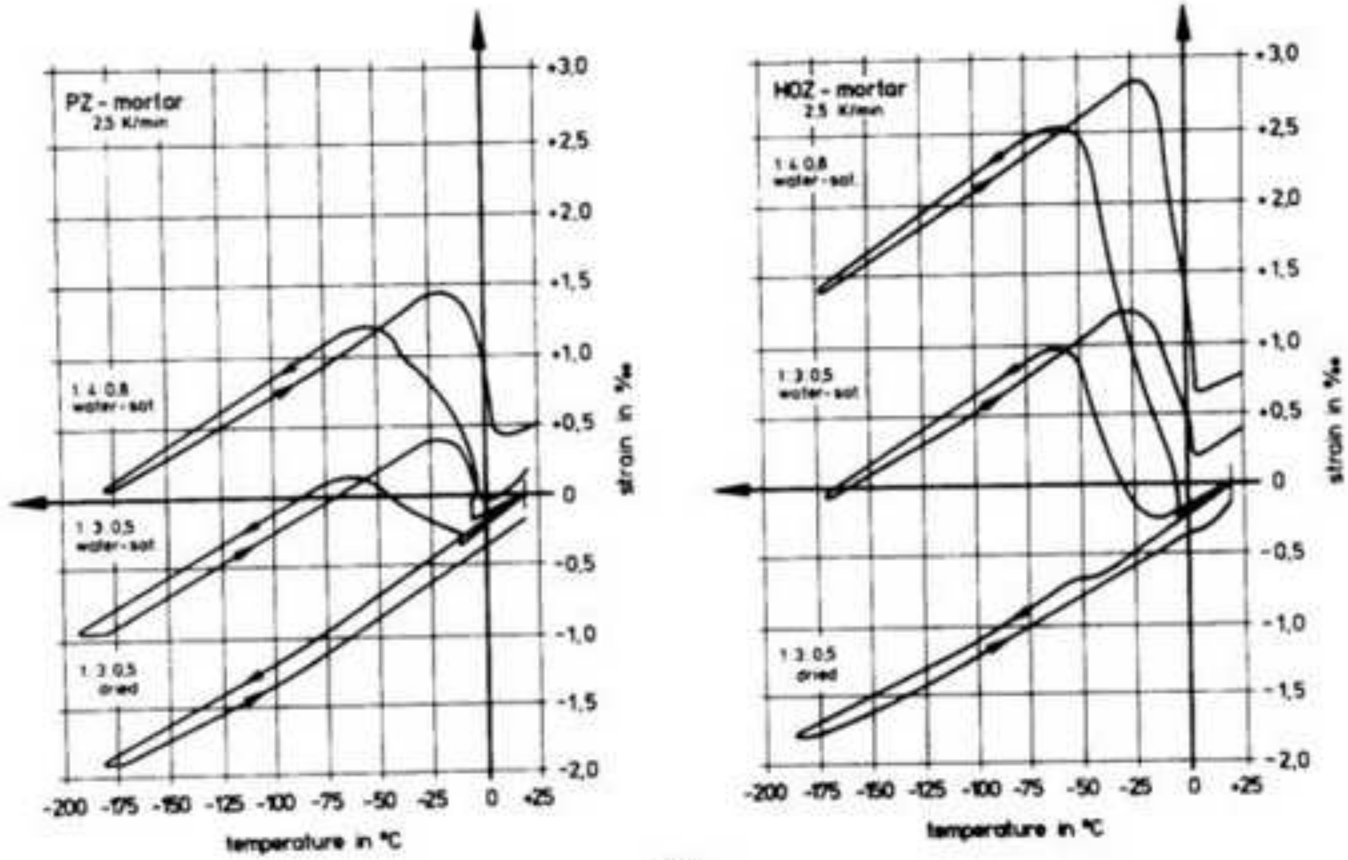

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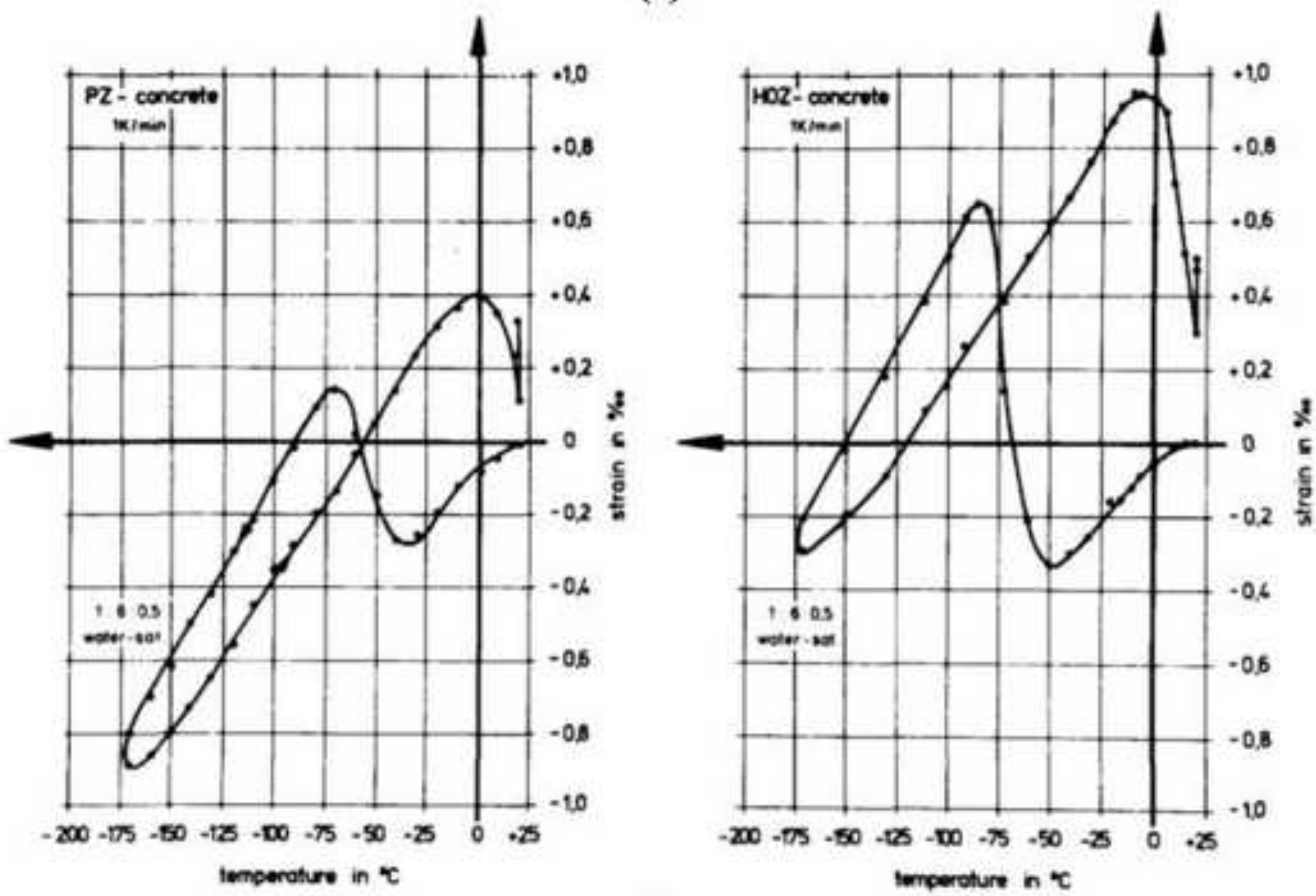

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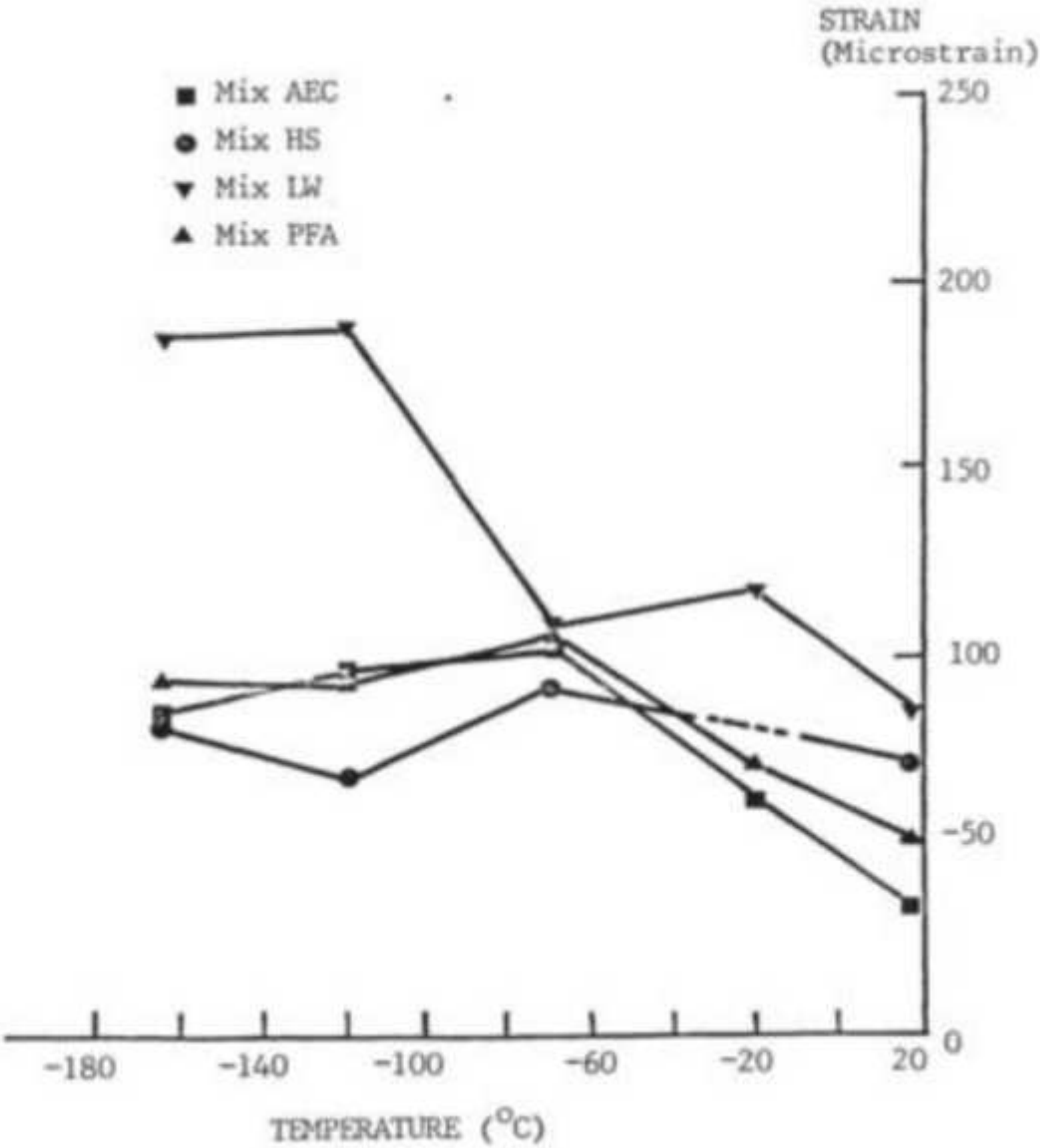

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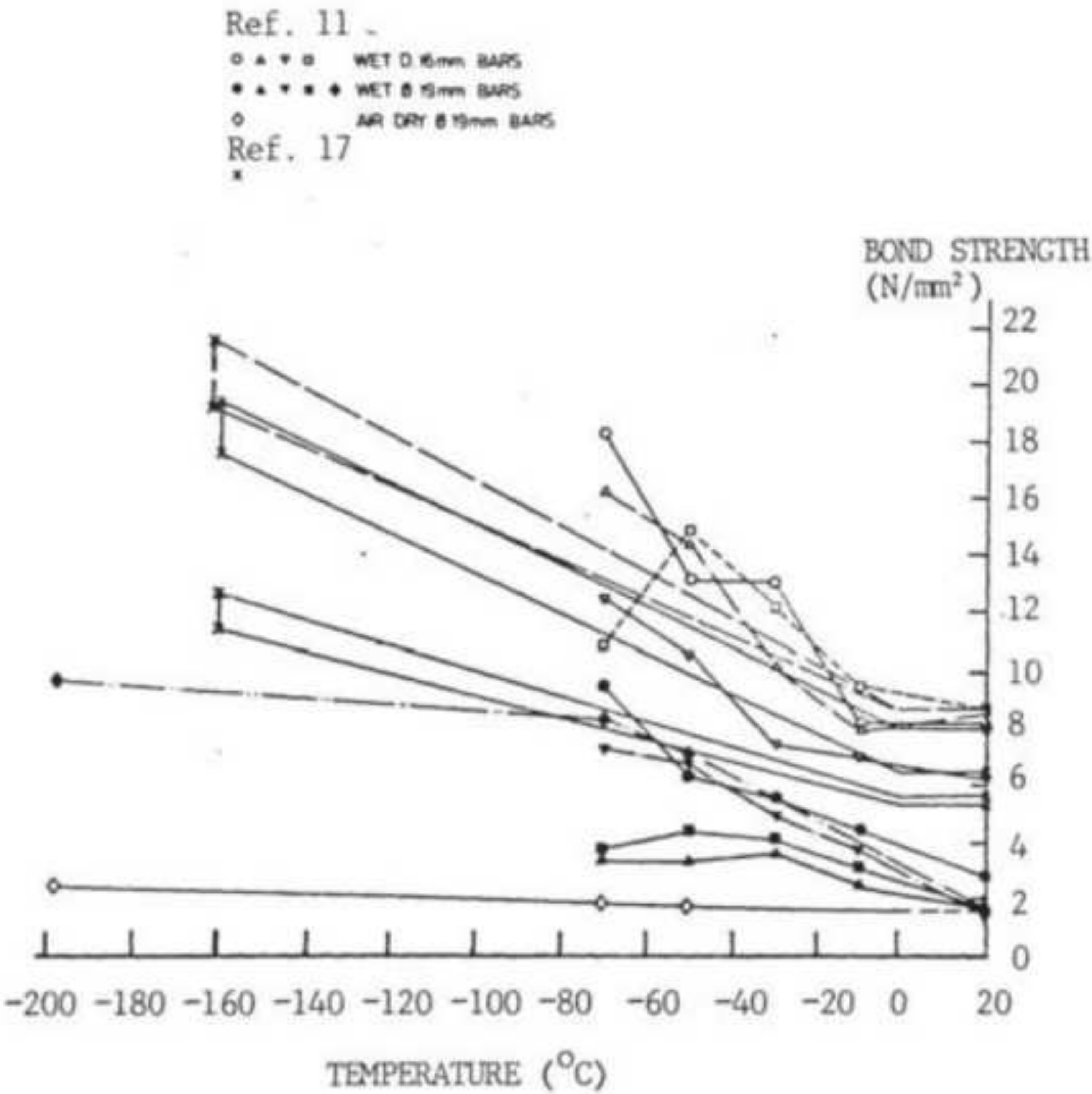

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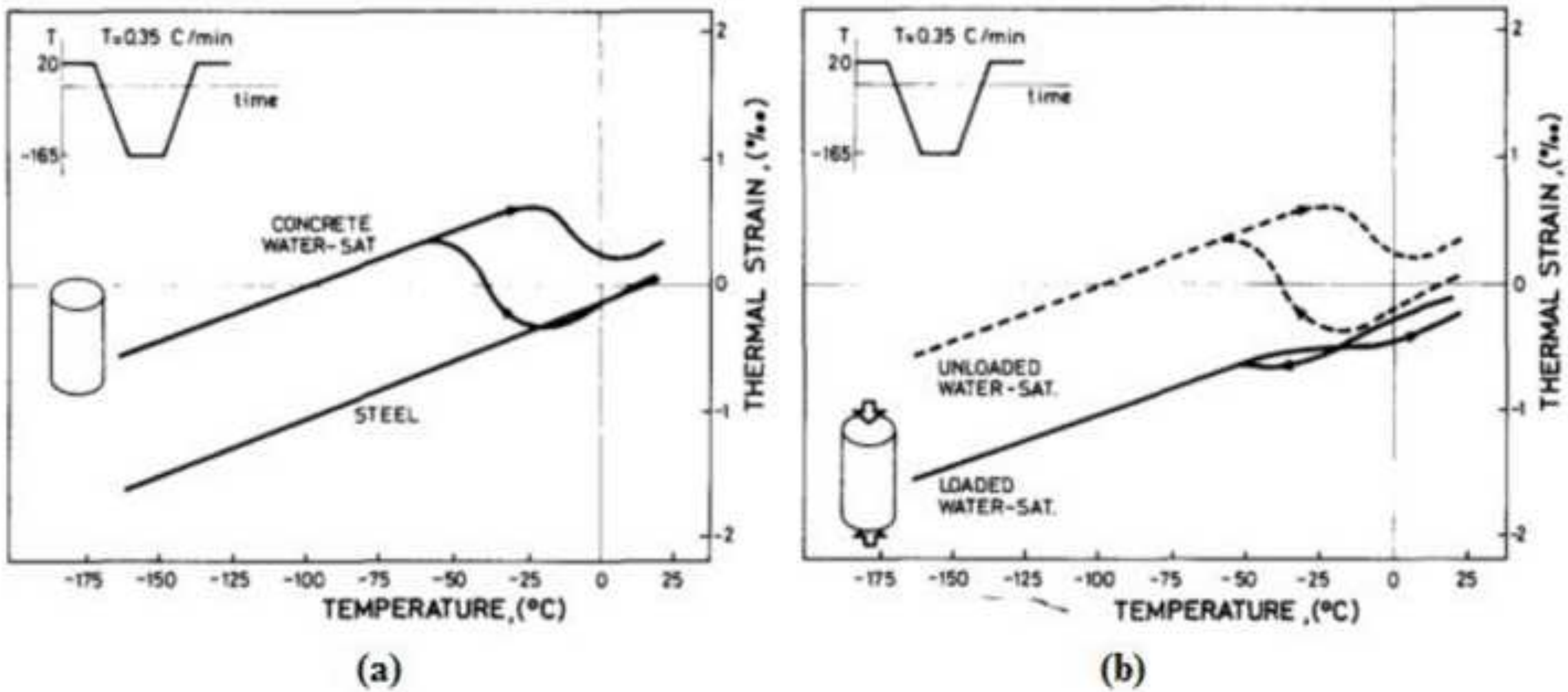

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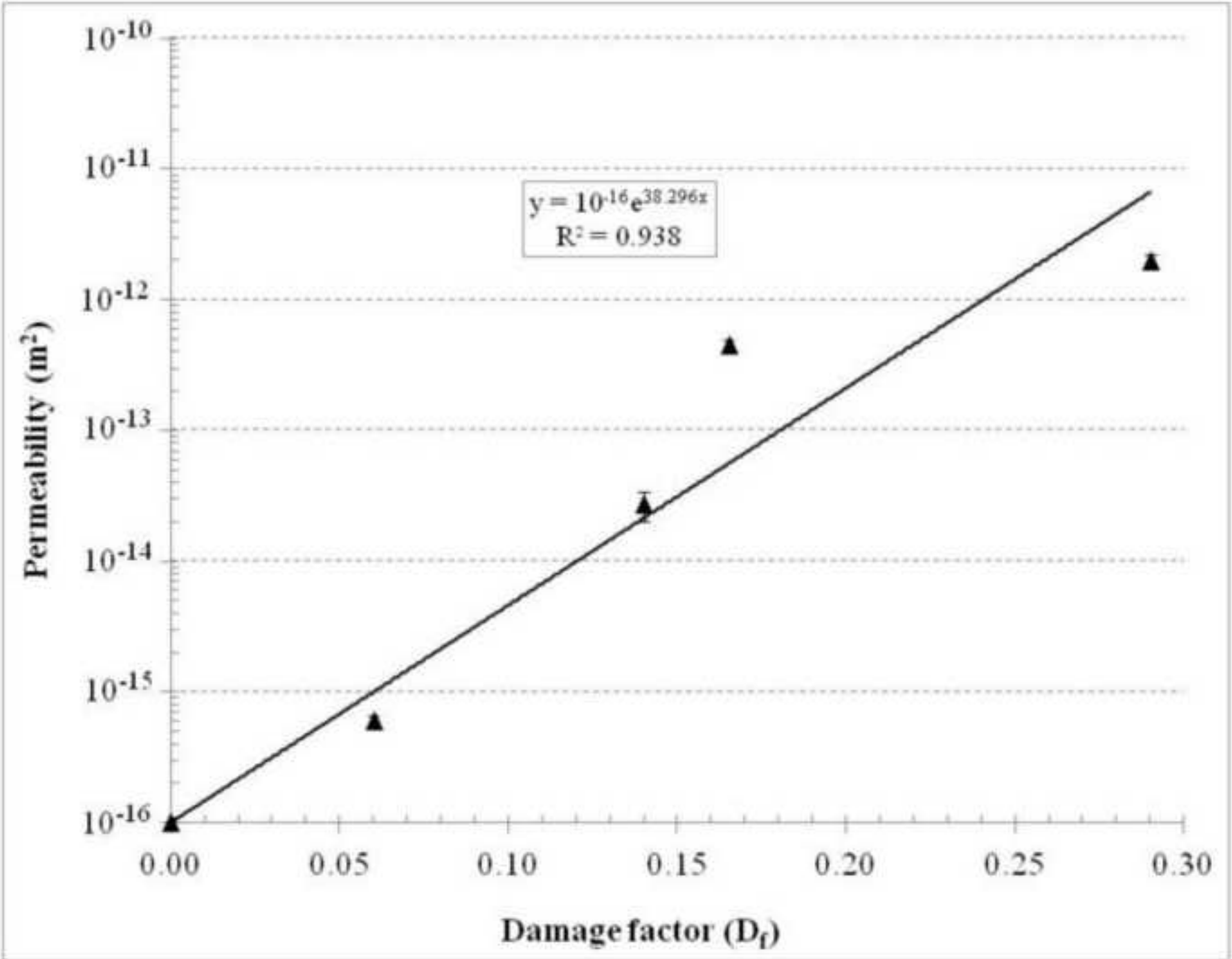

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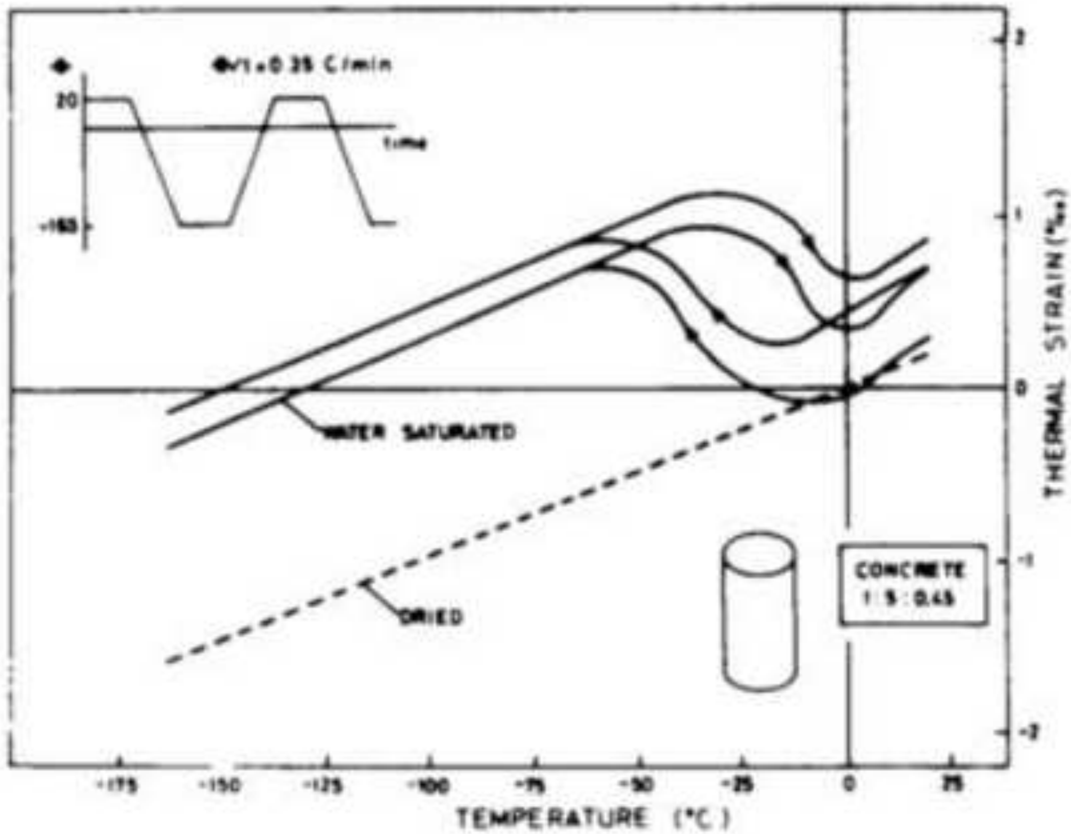

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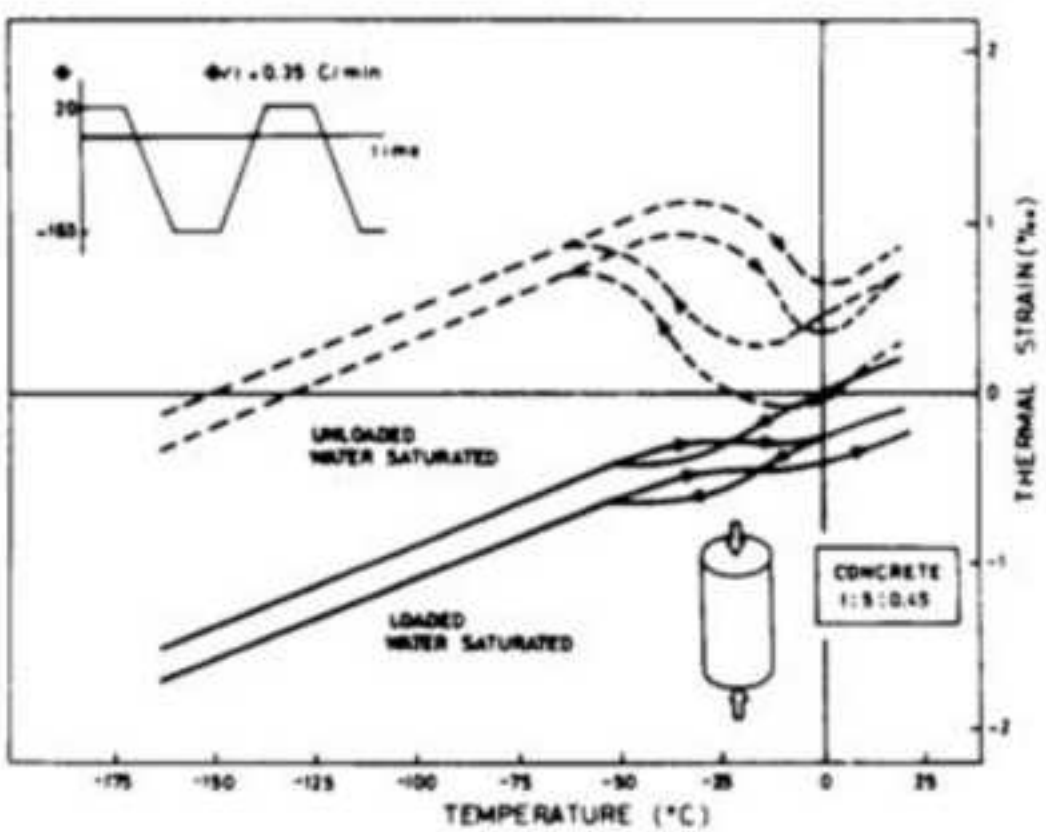

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