Influence of phytic acid on zinc phosphate cement

Jianguo Li, Sevald Forberg and Rune Söremark
Center for Dental Technology and Biomaterials, and Department of Clinical Research, Faculty of Odontology, Karolinska Institutet, Huddinge, Sweden

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The influence of phytic acid on the properties of zinc phosphate cement was studied by adding 2–13 wt% phytic acid to the liquid. Improved mechanical strength and stability were found for some cements prepared from commercial powders when liquids with increased phytic acid content were used. The results indicate that the formation of increased amounts of zinc phytate has a favorable effect on the properties of zinc phosphate cement. □ *Compressive strength; dental cements, chemistry; materials testing*

Jianguo Li, CDB, Box 4064, S-141 04 Huddinge, Sweden

Owing to its relatively high strength and easy mixing, zinc phosphate cement is commonly used as a luting agent in dentistry. The material has, however, certain disadvantages, such as transient pulpal irritation, brittleness, lack of adhesion to the tooth, and high water solubility (1–4).

The clinically effective life of zinc phosphate cement can be increased by reducing its water solubility. The strength of the cement is also an important property; lack of adhesion requires that it be retained by mechanical interlocking with surface irregularities of the tooth. Since the introduction of phosphate cement in dentistry almost one hundred years ago, numerous studies have been conducted to improve both the mechanical and physical properties and the bonding ability (5). Today, the development of zinc phosphate cement has advanced so that 70 MPa is defined as the lower limit for compressive strength and 2 mg P_2O_5/g as the upper limit for solubility in water (6). However, the introduction of phytic acid to the mixture may give additional improvements in these critical properties.

Phytic acid is the acid of myoinositol hexakis(dihydrogen phosphate) ($C_6H_6(H_2PO_4)_6$), a natural substance found in seeds. It can form stable complexes with metals, and it has been studied as a substitute for phosphoric acid in dental silicate and zinc oxide cements. The cements formed with phytic acid had lower water solubility and higher mechanical strength than the corresponding cements formed with phosphoric acid. However, the reaction between phytic acid and zinc oxide was so fast that the paste could not be used for dental applications (7).

To make practical use of the advantages of including phytic acid in zinc phosphate, the reaction can be modified by adjusting the content of phosphoric acid and other metals in the liquid. The purposes of this study were to define the optimum liquid composition and to evaluate the resulting properties of cements obtained by mixing the liquid with commercial cement powders.

Materials and methods

Phytic acid (Aldrich Chemical Co. Inc., USA) was supplied as a 40% (w/w) aqueous solution. Eleven experimental liquids were prepared with 44–57% orthophosphoric acid and 0–13% phytic acid and balanced with deionized water. Aluminum (aluminum foil GR, Merck), 0.5–3%, was added to the acid mixture to adjust the setting reaction. Table 1 shows the detailed compositions of these experimental liquids.

Table 1. Composition of experimental liquids

Code	Liquid composition (wt%)			
	H_3PO_4	Phytic acid	Al	H_2O
A	57	0	2.0	41.0
B	57	2	2.0	39.0
C	57	5	2.0	36.1
D	57	10	2.0	31.0
E	54	3	2.0	41.0
F	52	5	2.0	41.0
G	47	10	2.0	41.0
H	44	13	2.0	41.0
I	52	5	0.5	42.5
J	52	5	1.5	41.5
K	52	5	3.0	40.0
M*	—	0	—	—

* M = Phosphatine liquid.

The liquids were mixed with commercial cement powders on a glass plate. Three commercial powders were used: Phosphatine (Svedia, Sweden), Zinc (De Trey, Switzerland), and Harvard (Harvard, Germany). Eight samples were evaluated for each powder at each liquid composition for each factor. The ratio of powder to liquid was 2 to 1 by weight. The compressive strength, net setting time, water solubility, and film thickness of the specimens were measured in accordance with ISO 1566–1978 (E) (6).

Three-point bending strength of the three commercial cements, the powders mixed with their respective liquids, was compared with the strength of cements when the same commercial powders were mixed with experimental liquid K. The specimens, $25 \times 2 \times 2$ mm in size, were made in a removable stainless steel mold and kept in deionized water for 24 h at 37°C. The strength tests were performed in a universal testing machine (Alwetron, 50T) with a span of 22 mm at a crosshead speed of 1 mm/min. Ten specimens were used for each condition. All data were statistically evaluated by use of the *t* test.

Results

The compressive strengths obtained with Phosphatine powder and various experimental liquids are given in Table 2, together with the film thickness, setting time, and solubility. For liquids A through D, an increase of phytic acid (0–10%) and a decrease in water content (41–31%) delayed the setting reaction (5–12 min) and increased the mechanical strength (67–127 MPa).

The compressive strength of cements made from Phosphatine powder and liquids A and E through H was increased when phytic acid was present and decreased when the content of phytic acid was 10% or more. The influence on setting time in this group was not significant, except for liquid E.

The compressive strength of cements prepared from Phosphatine powder and liquids I, J, F, and K was increased (47–164 MPa) when the amount of aluminum was increased (0.5–3%) while the phytic acid content was held constant (5%). The highest strength was obtained at 3% aluminum. The viscosity of the paste increased with increasing concentrations of aluminum.

The water solubility of cements made from the liquids containing phytic acid was significantly reduced compared with those made from the liquids without phytic acid—that is, liquids A and M. Liquid E resulted in a cement with the lowest solubility, and liquids F, H, I, and J in cements with the highest solubility.

Fig. 1 shows the compressive strength of cements made with the original liquids and with liquids F and K. Strength was significantly increased ($P < 0.05$) when liquids K and F were used with all three cements, except when liquid F was used with the Harvard cement.

The three-point bending strength of the cements prepared with the commercial powders with the original liquids and with liquid K is shown in Fig. 2. The observed differences in strength were significant for Phosphatine and Zinc but not for Harvard.

Discussion

Prosser et al. (7) suggested that phytic acid has a multifunctional nature, with the potential of increase in cross-linking reactions and the formation of strong complex with poly-

Table 2. Properties of cements prepared of the experimental liquids and Phosphatine powder

	Film thickness (μm)	Setting time (min)	Compressive strength (MPa)	Solubility P_2O_5 (mg/g)
A	<25*	5.0	67 ± 17	3.90 ± 0.30
B	<25	9.9	112 ± 3	ND
C	<25	11.4	114 ± 11	ND
D	>40	12.0	127 ± 4	ND
E	<25	7.3	143 ± 18	0.05 ± 0.01
F	<25	5.2	145 ± 7	0.35 ± 0.10
G	<25	5.0	125 ± 9	0.18 ± 0.01
H	>40	5.0	132 ± 3	0.32 ± 0.10
I	<25	4.7	47 ± 6	0.35 ± 0.20
J	<25	5.3	69 ± 4	0.34 ± 0.10
K	<25	6.3	164 ± 15	0.16 ± 0.04
M	<25	7.3	112 ± 11	1.25 ± 0.13

ND = not determined.
* Film thickness maximum 25 μm for type I and 40 μm for type II according to ISO 1566–1978 (E).

valent cations. Compared with phosphate-bonded cements, phytate yields more stable and stronger cements. This may explain that the strength of cement B was much higher than that of cement A. As can be seen from series A through D in Table 1, when the concentration of phytic acid was increased from 0 to 2%, the compressive strength increased almost twofold, but little additional strength was gained when the phytic acid content was further increased up to 10%. Replacing phosphoric acid with phytic acid up to 13%, with other factors being held constant (series A and E-H), resulted in a maximum compressive strength in the region of 3–5% phytic acid. With a content of more than 5% phytic acid, the increase in paste viscosity resulted in more defects than

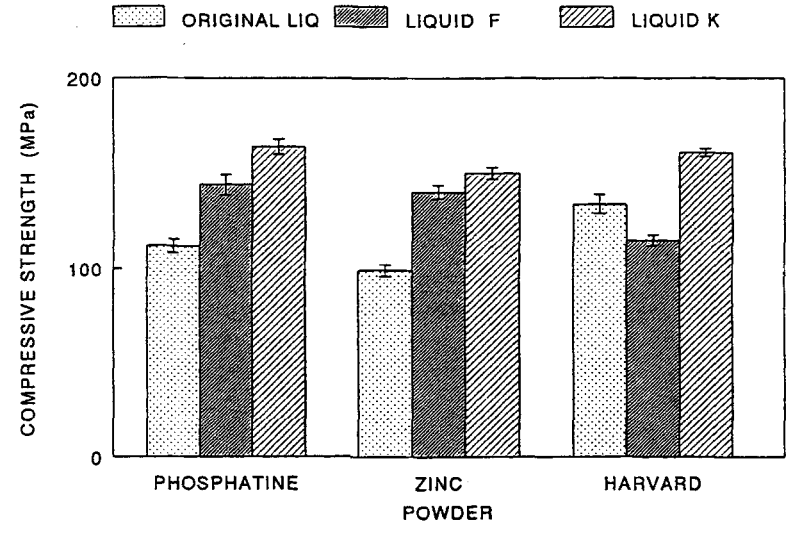

Fig. 1. Compressive strength of cements prepared from the original cement liquids and the experimental liquids. Error bars represent the standard error. Each column represents eight specimens.

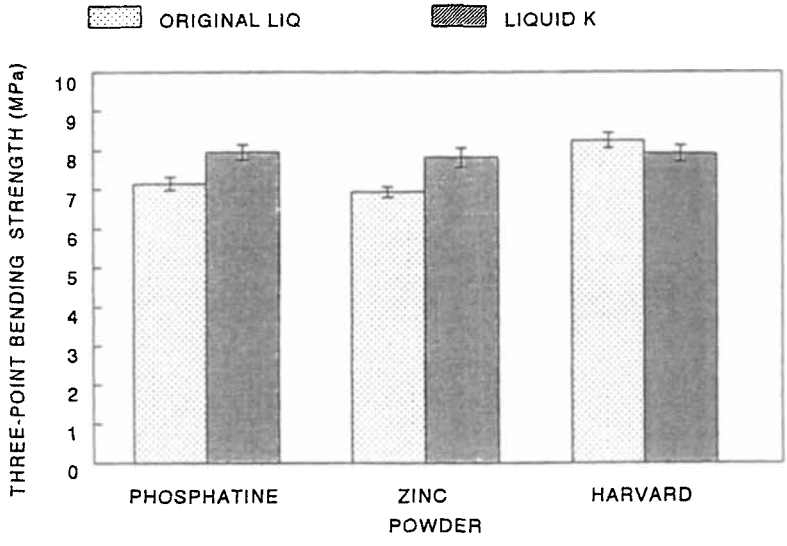

Fig. 2. Three-point bending strength of cements prepared from the original cement liquids and one experimental liquid. Error bars represent the standard error. Each column represents 10 specimens.

was seen in pastes with low viscosity, with a possible negative influence on strength.

It is obvious that aluminum is an important factor for both the strength and setting reaction of cement (8, 9). The optimal concentration of aluminum seems to be between 2% and 3% by weight in the liquid. Lower concentrations reduce the strength, and higher concentrations result in too high viscosity of the cement paste for dental application.

The cement-forming reaction of phytic acid alone is very rapid when mixed with the powder, and the setting time becomes short. From Table 2 it is obvious that a properly selected ratio of phytic acid and phosphoric acid can result in a suitable setting time. The increasing setting times in series A through D reflect the importance of water contents. Acceleration of the reaction by phytic acid has been more than counterbalanced by lower concentrations of water. When the concentration of water is constant, as in series E through H, the setting time may be held almost constant at the various concentrations of phytic acid (3–13%). However, within that range, pastes with concentrations of phytic acid above 5% become too viscous for dental application, and the film thickness increases with phytic acid above 10%.

Dupuis et al. (10) concluded that the dissolution of zinc phosphate cement was continuous and seemed to follow a diffusion law. In Table 2 it can be seen that the leach from zinc phosphate cement is reduced substantially by replacing some phosphoric acid with phytic acid. This may be because zinc phytate is much more stable than zinc phosphate (7). That is the main advantage of using phytic acid. Thus, the effective clinical life of the cement may be increased. This effect should be of special interest in some applications, such as in orthodontics, when a relatively large amount of cement is directly exposed to the saliva. The water solubility of cements prepared from liquids F, I, and J is the same when using the same concentration of phytic acid (5%) in these liquids. The explanation of the lower water solubility of cement made from liquid K may be related to the increased aluminum content (3%) in the liquid. However, at present it seems to be difficult to explain the differences between the liquids E and G and other liquids. The compressive strengths of the other two commercial cements (Zinc and Harvard) were improved by using our ex-

perimental liquids. Analyses showed that in all cases except one (Harvard powder and liquid F), significant increases were achieved ($P < 0.05$). The results suggest that, although the chemical compositions of cement powders vary (11), strength usually can be improved by using phytic acid. Three-point bending strength (Fig. 2) showed the same pattern as compressive strength. The decrease in compressive and bending strength of cement prepared from Harvard powder and liquid F is probably due to a content of polymer in Harvard liquid (5). This polymer seems to improve the strength of zinc phosphate more than that of phytic acid in liquid F.

In summary, an improved zinc phosphate cement can be obtained by adding a low percentage of phytic acid to the conventional cement liquid. The cement has an appropriate setting time and has greater strength and lower water solubility. Several other factors, such as free water, metal content, and acid concentrations, also have effects on the setting reaction and properties of the cements.

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