Al-imad Journal of Humanities and Applied Sciences (AJHAS)
مجلة العماد للعلوم الإنسانية والتطبيقية
ISSN: 3106-0323
Volume 2 – Issue 2 – 2026
Pages 538-547

Total Soluble Fluoride Derivatives Determination in Three Different Commercial Brands of Toothpaste in Malaysia Market by Using Fluoride Ion-Selective Electrode

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تحديد مشتقات الفلوريد الذائبة الكلية في أربع علامات تجارية تجارية لمعجون الأسنان في السوق الماليزية باستخدام القطب الانتقائي للأيونات

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Received: 29-06-2026; Accepted: 30-07-2026; Published: 20-08-2026

Abstract:
This study investigated and ranked the total soluble fluoride derivatives across four commercial toothpaste brands available in the Malaysian market using the ion-selective electrode (ISE) technique. Accurately quantifying soluble fluoride is essential for evaluating the anticariogenic efficacy and safety profiles of dental hygiene products. Experimental procedures involved sample preparation, digestion, and potentiometric determination against standardized fluoride solutions. The findings revealed notable variations in total soluble fluoride concentrations among the tested brands. Specifically, the Sensodyne sample exhibited the highest concentration of total soluble fluoride derivatives at 3524.2897 ppm, followed by Darlie at 1864.9310 ppm and Zact at 1509.0499 ppm. Conversely, Colgate recorded the lowest total soluble fluoride derivatives at 1206.4733 ppm. These variations correlate directly with the respective formulation characteristics, abrasive percentages, and chemical compositions of each brand. The results provide critical data regarding commercial fluoride availability, assisting consumers and dental professionals in selecting appropriate products that balance effective caries prevention with safety against dental fluorosis.

Keywords: Fluoride, Ion-Selective Electrode, Toothpaste, Malaysia, Soluble Fluoride Derivatives, Potentiometry.

الملخص
تناولت هذه الدراسة تحديد وترتيب إجمالي مشتقات الفلوريد الذائبة في أربع علامات تجارية تجارية لمعجون الأسنان متوفرة في الأسواق الماليزية باستخدام تقنية القطب الانتقائي للأيونات .*(ISE)* يُعد القياس الكمي الدقيق للفلوريد الذائبي أمراً بالغ الأهمية لتقييم الفعالية المضادة للتسوس ومعايير الأمان لمنتجات العناية بالأسنان. تضمنت إجراءات العمل تحضير العينات، والهضم الكيميائي، والقياس الجهدد للمحتوى مقابل محلول الفلوريد القياسي. أظهرت النتائج تفاوتاً ملحوظاً في تركيزات إجمالي الفلوريد الذائبي بين العلامات التجارية المختبرة؛ حيث سجلت عينة *(Sensodyne)* أعلى تركيز لإجمالي مشتقات الفلوريد الذائبة بواقع 3524.2897 جزء في المليون، تليها *(Darlie)* بواقع 1864.9310 جزء في المليون، ثم *(Zact)*بواقع 1509.0499 جزء في المليون، بينما سجلت علامة *(Colgate)* أدنى قيمة بإجمالي مشتقات الفلوريد الذائبة بواقع 1206.4733 جزء في المليون. ترتبط هذه الفروق ارتباطاً مباشراً بخصائص

التركيبة، والنسب المئوية للمواد الكاشطة، والتركيب الكيميائي لكل علامة تجارية. توفر هذه النتائج بيانات جوهرية حول مدى توفر الفلوريد التجاري، مما يساعد المستهلكين وأخصائيي الأسنان في اختيار المنتجات المناسبة التي تحقق التوازن بين الوقاية الفعالة من تسوس الأسنان والسلامة من التسمم بالفلور.

الكلمات المفتاحية: الفلوريد، القطب الانتقائي للأيونات، معجون الأسنان، ماليزيا، مشتقات الفلوريد الذائبة، القياس الجهدي.

INTRODUCTION

Fluoride is the 13th most abundant element on the earth's crust and it is a chemical ion of the element fluorine. Fluoride is found naturally in soil, water, foods, and several minerals, such as fluorapatite and fluorite. Fluoride has both beneficial and detrimental effects on human health. In terms of dental health, the prevalence of dental caries is inversely related to the concentration of fluoride in drinking water; while there is a dose-response relationship between the concentration of fluoride in drinking water and the prevalence of dental fluorosis. In terms of general health, in communities where drinking water and foodstuffs are excessively high in fluoride, skeletal fluorosis and bone fracture are the most relevant adverse effects (Lennon et.al, 2004).

Toothpastes containing fluoride make up more than 95% of all toothpaste in the market. Investigations into the effectiveness of adding fluoride to toothpaste have been carried out since 1945 and cover a wide range of active ingredients in various abrasive formulations. Fluoride compounds and their combinations which have been tested for the control of dental decay include sodium fluoride, stannous fluoride, sodium monofluorophosphate and amine fluoride. It is well recognized that the decline in the prevalence of dental caries recorded in most industrialized countries over the past 30 years can be attributed mainly to the widespread use of toothpaste that contain fluoride (Oral Health Services Research Centre,1999). The amount of fluoride contained in fluoride toothpaste normally is indicated on the toothpaste tube. Fluoride content in a toothpaste can be expressed in percent of weight of fluoride over volume of toothpaste (% w/v), percent of weight of fluoride over weight of toothpaste (% w/w) and parts per million of fluoride (ppm F). The most efficient and the most widely used of unit to inform the public of the amount of fluoride in toothpaste is ppm F (Oral Health Services Research Centre, 1999). The Institute of Medicine, Food and Nutrition Board has estimated that the tolerable upper limit for human daily intake of fluoride for adults and children over 8 years of age 19 is 10 mg per day. Ten independent U.S and Canadian studies published from 1958 to 1987 showed that dietary fluoride intakes by adults ranged from 1.4 to 3.4 mg/day in areas where the water fluoride concentration was 1.0 mg/L. Meanwhile, in area where the water concentration was less than 0.3 mg/L, daily intakes ranged from 0.3 to 1.0 mg/day (F. Pollick, 2004). There are various methods of analysis to measure fluoride levels such as ion selective electrode (ISE), spectrophotometer and gas chromatography. However, ISE method has found a wide application area because of being a simple and practical technique (Speirs, 1983). There are several types of ISE such as Glass membrane, Solid-state electrode, Liquid-based electrode and Compound electrode. ISE techniques offer several advantages such as linear response, over 4 to 6 orders of magnitude of Absorbance, non-destructive technique where no consumption of analyte, non-contaminating, short response time and unaffected by color or turbidity. Thus, in this study the measurement of fluoride was performed by the Solid-state electrode ion-specific electrode method as well.

METHODS AND MATERIALS

Apparatus

- 500 m, 250 ml and 100 ml beakers
- Thermometer

- 5000 µL micropipette
- 10 ml pipette
- 500 ml and 250 ml volumetric flasks
- Stop watch

Chemicals

- Anhydrous sodium chloride
- Sodium chloride
- Sodium citrate
- Glacial acetic acid
- Sodium hydroxide
- De-ionized water

Instruments

- Thermo Scientific Orion VERSA STARTM pH/ISE/Temperature Module

- Analytical balance

- Sonicator
- pH Meter

Sampling

A total of 4 samples of different brands of toothpaste were analyzed in this experiment. All samples are bought from available in market.

i. Sensodyne
ii. Darlie Double Action
iii. Colgate Anti-cavity toothpaste
iv. Zact whitening toothpaste

Each sample was prepared triplicate and each of it was read 3 times.

Preparation of TISAB

The total ionic strength adjustment buffer (TISAB) was prepared in 500 ml of volumetric flask by mixing 28.5 ml of glacial acetic acid, 29 g of sodium chloride, 0.15 g of sodium citrate and then mark up with water to the mark. Its pH was adjusted to 5.3 with sodium hydroxide and then diluted to the mark with distilled water.

Standard of Fluoride Preparation in water

Stock solution of 500 ppm was prepared by weighing 0.5525 g sodium fluoride by using analytical balance. Then, transfer to 500 ml volumetric flask and finally diluted to the mark with water. Prior to the next steps, the standard was put in sonicator for 4 minutes. Standard solution of 200 ppm was then obtained by diluting with water 40 ml stock solution in 100 ml of volumetric flask. Standard solution of 100 ppm was then obtained by diluting with water 50 ml of 200 ppm of fluoride standard in100 ml of volumetric flask. Standard solution of 50 ppm was then obtained by diluting with water 50 ml of 100 ppm of fluoride standard in 100 ml of volumetric flask. Standard solution of 20 ppm was then obtained by diluting with water 40 ml of 50 ppm of fluoride standard in 100 ml of volumetric flask. Standard solution of 10 ppm was then obtained by diluting with water 50 ml of 20 ppm of fluoride standard in 100 ml of volumetric flask. . Standard solution of 2 ppm was then obtained by diluting with water 20 ml of 10 ppm of fluoride standard in 100 ml of volumetric flask. Finally, Standard solution of 1 ppm was then obtained by diluting with water 50 ml of 2 ppm of fluoride standard in 100 ml of volumetric flask.

Standard of Fluoride Preparation in water

The preparation of a series of fluoride standard in TISAB were using the standard prepare in water before. 250 ppm, 100 ppm, 50 ppm, 25 ppm, 10 ppm, 5 ppm, 1 ppm and 0.5 ppm of fluoride working standard in TISAB was prepared by pipette 10 ml of 500 ppm, 200 ppm, 100 ppm, 20 ppm, 20 ppm, 10 ppm, 2 ppm and 1 ppm of fluoride standards in water respectively into beaker and added 10 ml of TISAB.

Sample Preparation for total soluble fluoride derivatives in toothpaste

Few centimeters of toothpaste extruded from primary container was rejected as a precaution for contamination. 2 gram ±0.05 of sample was weighed and transferred to plastic tube. Then 10 ml of water was added. The sample was then put in sonicator about 4 minutes and stir continuously with plastic rode. After that, the sample was filtered by using 0.45 µm nylon syringe filter. 1 ml of filter sample was then mixed thoroughly with 1 ml of Hydrochloric acid (HCl) and seals it. Then the sample was incubated at 25 - 30ºC for 1 hour. After that, accurately 8ml of water was added to the sample solution. Then 5 ml of it is transfer to another plastic tube and was added with 1ml of 4M Sodium Hydroxide (NaOH) and finally added 6 ml of TISAB. Repeat all procedures for other two replicates samples and another three brands of toothpaste.

Prior to the analysis with ISE, the temperature of the sample was ensuring within ± 1 ºC of the average temperature measured previously for the fluoride standards in TISAB. The fluoride ion elctrodes was immersed in the solution, and magnetic stirrer was continuously stirring the sample till the mV reading is stable. By reference to the calibration curve, the fluoride concentration in test solution was calculated. The soluble fluoride derivatives in the toothpaste calculated by equation below:

$F_d = 100E (1 + 5/D – C/100)$

F_d = Soluble fluoride derivatives in the toothpaste.

E = Concentration of Fluoride ion in a solution.

D = Mass of toothpaste in sample in gram.

C = Insoluble material in toothpaste expressed in percentage.

Procedure for insoluble material in toothpaste

Few centimeters of toothpaste from primary container were rejected. 2 gram of toothpaste was transferred into weighed plastic tube. Then 10 ml of distilled water was added to the samples and stir with plastic rod till homogenize. The sample was then centrifuged with 6000 RPM speed for 10 minutes. After centrifugation process the insoluble material will accumulated at the bottom of the tube, upper layer of clear supernatant was rejected with Pasteur pipette. The steps were repeated twice to ensure all the soluble material is completely removed. The tube with remaining insoluble material was then placed in oven at 60 degree for 24 h. After that, cooled the tube and reweight to obtain the insoluble material in toothpaste from following equation:

C=100 X A/B

C = Insoluble material expressed in percentage

A = Mass of toothpaste of the sample in gram

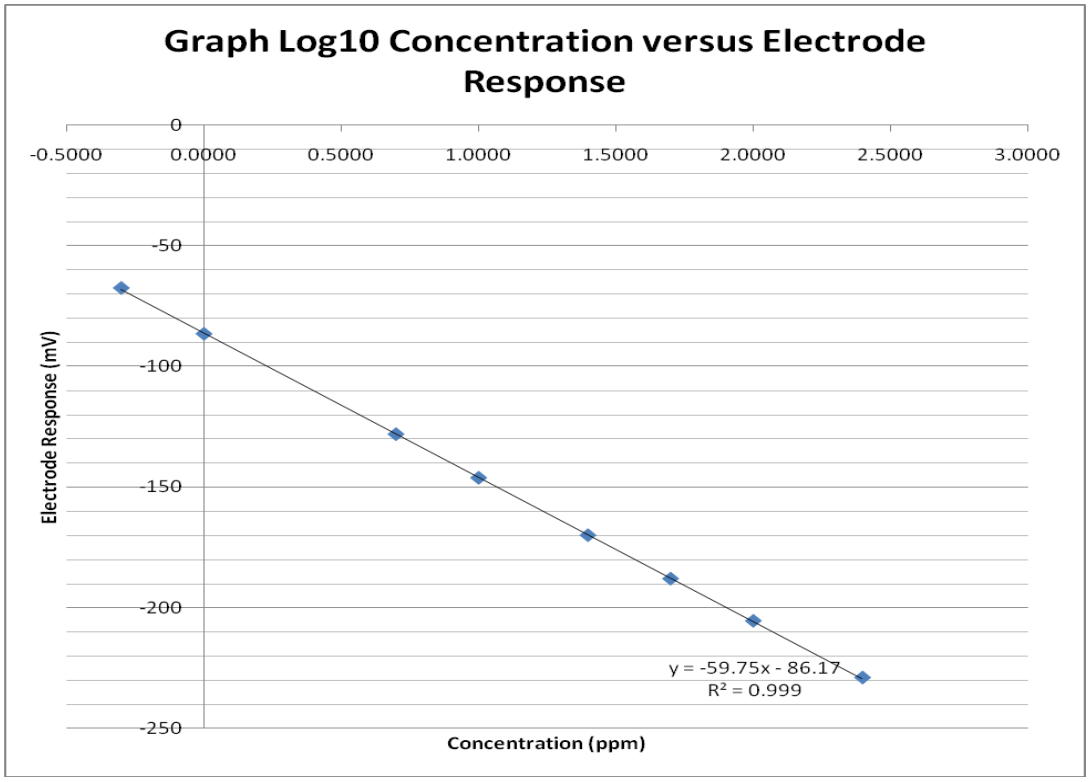
B = Mass of dried insoluble sample in gram

RESULTS AND DISCUSSION

The electrode response values that were recorded for each of the standard calibration solutions used in the direct Potentiometry technique are shown in Table 1 below.

Table 1: The electrode response values of the fluoride standard solutions.

Concentration of F^- (ppm)	Log10 Concentration	Electrode Response (mV)
0.5	-0.3010	-67.5
1.0	0.0000	-86.5
5.0	0.6990	-128.1
10.0	1.0000	-146.2
25.0	1.3979	-170
50.0	1.6990	-188
100.0	2.0000	-205.5
250.0	2.3979	-229



Graph 1: Calibration Standard of Fluoride using ISE.

Prior to the standard fluoride analysis by ISE, pure TISAB was analysing first, assuming the TISAB is free from Fluoride ion. The TISAB act as a blank of the analysis and the mV of the electrode reading was Re-zero. So, the samples with no fluoride content should have mV=0. Instrument calibration was build up from the data from the standards. The graphs establish the relationship between the electrode response and the accepted values of the calibration standards. Normally, the relationship is expressed in a graph as linear equation. The equation was used estimates of the amount of fluoride present in unknown samples (Barwick, 2003). The Coefficient Determination, R^2 is related to linearity of the results. The linearity is the ability to obtain test results that are directly proportional to the concentration of an analyte in the sample. This may be true within a given range. Once the calibration curve is set up, the linearity of this curve is tested. The linearity for each analyte is assessed by checking the performance of recovery throughout the stated range of the testing. The value of R^2 when more than 0.90 indicate the calibration curve is reliable. As the calibration curve in this study is obtained at R^2 value is 0.999, this show that all the F^- standards that are prepared were reliable standards (Jinadasa 2010).

Table 2: Electrode response values of the sample solutions with the calculated concentration values

Toothpaste Samples	Weight of the Samples (g)	Electrode Response(mV)			
		1	2	3	Average
Sensodyne-1	4.0172	-165.8	-165.8	-165.8	-165.8
Sensodyne-3	2.0075	-147.4	-147.1	-146.4	-147.0
Sensodyne-3	2.0005	-148.2	-146.9	-147.7	-147.6
Mean					-153.47

Darlie-1	2.0340	-135.9	-135.9	-135.9	-135.9
Darlie-2	2.0218	-131.6	-131.9	132.1	-131.9
Darlie-3	2.0251	-133.6	-133.0	-133.8	-133.5
Mean					-133.7
Zact-1	2.0433	-130.9	-131.4	-131.4	-131.2
Zact-2	2.0670	-126.2	-127.8	-127.9	-127.3
Zact-3	2.0541	-126.9	-126.9	-126.3	-126.7
Mean					-128.40
Colgate-1	2.0279	-111.4	-113.2	-114.2	-112.9
Colgate-2	2.0178	-132.8	-132.6	-133.2	-132.9
Colgate-3	2.0394	-119.7	-119.4	-119.6	-119.6
Mean					-121.80

The outlier test was performed for Sensodyne data, Sensodyne-1 was expected as outlier of the result. Grubbs' test was used at 95% confidence level.

Sensodyne Data: -165.8, -147.6, -147.0

Hypothesis

H_0: -165.8 is not outlier

H_a: -165.8 is outlier

1) **Test Statistical**

$$G = \frac{|\text{Suspected Value} - \text{X bar}|}{s}$$

G = 165.8 – 153.47 / 10.68

= 1.15

2) **Critical value**

At 95% confidence level

$N = 3$ $G_{a/2} = G_{0.025} = 1.153$

3) **Rejection region**

Reject H_0 if $G > G_{0.025}$

4) **Interpretation**

The G value< then $G_{0.025}$, failed to reject H_o, -165.8 is not an outlier.

Table 3: Show the average electrode response of 4 different brands of toothpaste and their concentration fluoride ion derived from equation obtain from Calibration graph.

Toothpaste Samples	**Average (mV) (1st sample + 2nd sample +3rd sample)/3, Y**	**Log10 Concentration of Fluoride ion in solution X=(Y + 86.17)/-59.75**	**Concentration of Fluoride ion in solution (ppm), E E=Antilog X**
Sensodyne	-153.47	1.1264	13.3770
Darlie	-133.77	0.7967	6.2611
Zact	-128.40	0.7068	5.0907
Colgate	-121.80	0.5963	3.9475

The concentration of fluoride ion in samples solution was determined by information from calibration graph. Log10 Concentration of fluoride ion in samples were obtained by deriving the linear equation of calibration graph, Y= -59.75X – 86.17. The highest concentration of fluoride ion is in Sensodyne samples, followed by Darlie, Zact and the lowest was Colgate which is 13.3770 ppm, 6.2611 ppm, 5.0907 ppm and 3.9475 ppm respectively. Fluoride ion prevents teeth decay in two main ways. First, it reduces the ability of bacteria in plaque to generate tooth-weakening acids, and it helps re-mineralize those tooth areas where acid attack has already begun. Just assuming the fluoride ion only given beneficial to human, Sensodyne is the most effective toothpaste for preventive of teeth decay followed by Darlie, Zact and the lastly Colgate. But excessive fluoride may lead to dental fluorosis. As low level and excessive level of fluoride gives possibilities of negative effects to teeth health, led the study to determine highest level intake of fluoride ion in human with no risk to human health (F.Pollick, 2004). According to the U.S. Department of Agriculture, the Dietary Reference Intakes, which is the "highest level of daily nutrient intake that is likely to pose no risk of adverse health effects", specify 10 mg/day for most people. Every human lifestyle is different, how many times the individual brush his teeth and other source of fluoride in his daily routine should be considered for determining which types of toothpaste is suitable for him.

Table 4: Shows the insoluble material expressed in percentage of 4 different brands of toothpaste

Brands	Mass of toothpaste of the sample in gram (A)	Mass of dried insoluble sample in gram (B)	Insoluble material expressed in percentage C=100 X A/B
Sensodyne	**2**	0.469	**23.45**
Darlie	**2**	0.9768	**48.81**
Zact	**2**	0.938	**46.90**
Colgate	**2**	0.8174	**40.87**

The insoluble material represents in the samples also known as abrasives. These insoluble particles help remove plaque from the teeth. The removal of plaque and calculus helps minimize cavities and periodontal disease. The common abrasives used include Dicalcium Phosphate Dihydrate, insoluble Sodium Metaphosphate, Calcium Pyrophosphate, Calcium Carbonate, Alumina Trihydrate, Magnesium Trisilicate and more recently is silica gels. These abrasives have a hardness ranging between that of dentine and a value below that of enamel in order to be safe but effective stain removers. Nevertheless, cervical tooth abrasion can occur, creating wedge-shaped notches usually near the gingival margin. In severe cases, Pulpal Pathosis and Periapical lesions may result (C. Reynolds, 1994). Hence the selection of toothpaste for individual depends on the plaque and calculus, individual with high amount of plaque or calculus should used toothpaste containing high amount of abrasives. A highest abrasive was

sample from Darlie followed by Zact, Colgate and the lowest is Sensodyne which is 48.81%, 46.90%, 40.87% and lastly 23.45% respectively. Sensodyne has the lowest abrasive suitable as what's being claimed, it is suitable for sensitive teeth. An abrasive is dental polishing agents which cause a small amount of enamel erosion which is termed "polishing" action. Sensitive teeth should use fewer amounts of abrasives to reduce or control the polishing action.

Table 5: Shows the total soluble fluoride derivatives in the toothpaste through calculation using formula as stated in procedures.

Brands of Toothpaste	**Insoluble material in toothpaste expressed in percentage, C=100 X A/B**	**Mass of toothpaste in sample in gram, D.**	**Concentration of Fluoride ion in solution (ppm), E Y=-59.75X – 86.17**	**Soluble fluoride derivatives in the toothpaste (ppm). Fd = 100E (1 + 5/D – C/100)**
Sensodyne	23.45	2.6751	13.377	3524.2897
Darlie	48.81	2.0270	6.2611	1864.9310
Zact	46.9	2.0548	5.0907	1509.0499
Colgate	40.87	2.0284	3.9475	1206.4733

The aimed of this study is to find the total soluble fluoride derivatives in toothpaste. In order to get it, the insoluble material in toothpaste and the concentration of fluoride ion in solution should identify first. After this parameter had identified, the total soluble fluoride derivatives in toothpaste was calculated by using formula stated in procedures, Fd = 100E (1 + 5/D – C/100). Highest amount of soluble fluoride derivatives is in Sensodyne sample which is 3524.2897 ppm, followed by Darlie-1864.9310, Zact – 1509.0499 and the lowest total soluble fluoride derivative is in Colgate which is 1206.4733 ppm.

The study has limitation which may cause the data is questionable. The apparatus available in labarotary is very limited. The study of fluoride ion should conduct in all plastic apparatus as the fluoride may react with the glass which cause the fluoride ion obtained may low from the true value. The centrifuge was not available in laboratory, the nylon filter was used to replace the centrifuge, but there is possibility the loss of fluoride ion during filtering process.

CONCLUSION

As the conclusion, the objectives were achieved. The Concentration of total soluble Fluoride Derivatives 4 different brands was identified and ranked. Highest concentration of soluble fluoride derivatives is in Sensodyne sample, followed by Darlie, Zact and the lowest total soluble fluoride derivative is in Colgate.

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APPENDICES

Stability study of TISAB and Standard Fluoride

Concentration of F ion	Log10 Concentration of F ion	21-Nov (mV)	28-Nov (mV)
TISAB		50.1	68.6
0.5000	-0.3010	-67.5	-76.7
1.0000	0.0000	-86.5	-98.7
5.0000	0.6990	-128.1	-140.1
10.0000	1.0000	-146.2	-157.6
25.0000	1.3979	-170	-181.6
50.0000	1.6990	-188	-199.8
100.0000	2.0000	-205.5	-217.6
250.0000	2.3979	-229	-241.6

The standard of fluoride prepared on 21 Nov 2014, was read again in 28 Nov 2014. The reading is slightly different.

Date	14-Nov	21-Nov	28-Nov
TISAB (pH)	5.20	5.16	5.10

The TISAB prepared on 14 Nov was read again prior to used it in analysis, the pH is slightly different but still in accepted range to used.

