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EXPERIMENTAL OBSERVATIONS ON THE HARDNESS OF ENAMEL

Thesis submitted to the
University of Sydney
for admission to the degree of
Doctor of Dental Science.

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"The search for truth in the domains of history and philosophy is carried on in constantly repeated endless duels between the sense of reality of the one and the inventive imaginative power of the other. The argument from facts is never able to obtain a definite victory over the skillfully produced opinion. How often does what is reckoned as progress consist in a skillfully argued opinion putting real insight out of action for a long time!

Now I was suddenly in another country. I was concerned with truths which embodied realities, and found myself among men who took it as a matter of course that they had to justify with facts every statement they made.

Intoxicated as I was with the delight of dealing with realities which could be determined with exactitude, I was far from any inclination to undervalue the humanities. On the contrary. Through my study of chemistry, physics, zoology, botany and physiology I became more than ever conscious to what an extent truth in thought is justified and necessary, side by side with truth which is merely established by facts."

Albert Schweitzer.

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INTRODUCTION

Mineralization and its converse, demineralization are two processes of intimate concern to the dental profession. Both processes have been studied extensively and both have been shown to involve a change in the hardness of the tissue whether it be bone, dentine or enamel.

The nature of the mineralization process is still in doubt (Neuman and Neuman, 1958). Attempts to explain calcification by solubility product relationships have not been successful, and the original phosphatase theory of Robison has been discarded. Cellular glycolysis which seemed to directly participate in the mineralization process was later suggested as a modification of the original theory. More recently, a seeding mechanism or epitaxy has replaced the precipitation concept. A good deal of evidence has been accumulated to show that, as with bone, acid mucopolysaccharides are involved in the calcification of enamel and dentine and that they may act as a template for the crystallization of the apatite structure (Irving, 1957). Whatever the final explanation may be, studies (Deakins, 1942; Avery and Visser, 1956, 1960) have shown there is a marked increase in enamel hardness during calcification. A study of enamel hardness may therefore throw some light on the calcification problem.

Dental caries is a disease rempant in modern civilized man. It attacks the preschool child soon after the eruption of the deciduous teeth. It becomes exceedingly prevalent among the school population and young adults and persists into old age. Finn (1952), in a comprehensive survey of the literature found that the percentage of children showing evidence of caries varies from 70 to 100 percent, depending upon the age of the children and the geographic location. In New South Wales, 99 percent of children 12 years of age, examined in a recent survey (Barnard, 1956) were affected by caries of the permanent teeth.

Three theories of dental caries attack have been evolved, namely the chemico-parasitic, the proteolytic and the proteolytic-chelation theories. The chemico-parasitic theory, initially enunciated by Miller (1890), still attracts the most adherents. Miller wrote as follows: "Dental decay is a chemico-parasitic process consisting of two distinctly marked stages: decalcification, or <u>softening</u> of the tissues, and dissolution of the <u>softened</u> residue. In the case of enamel, how ever, the second stage is practically wanting, the decalcification of the enamel signifying its total destruction." Miller recognized that softening or a decrease in hardness of the enamel was a direct result of decay. Simply stated this theory considers that the enamel is progressively destroyed by means of acids, almost certainly organic, which result from the

fermentation by oral organisms of suitable food residues retained in the mouth.

The proteolytic theory of caries differs from the above in emphasizing the protein elements of the enamel as the object of primary attack. The proteolytic-chelation theory proposes that bacteria or bacterial products complex with and remove the calcium salts in the absence of acid. All the theories agree, however, that softening is an integral part of the ultimate caries process. Indeed, softening of the enamel is the most widely used clinical criterion in the diagnosis of carious lesions.

This thesis endeavours to lead to a better understanding of the hardness properties of sound and fluorosed enamel and of the softening of enamel under standardized conditions. The experimental studies in this dissertation have contributed new information which it is hoped can be applied to the basic problem of calcification and decalcification as outlined in the introduction.

Chapter I contains a detailed review of the available literature on the hardness of enamel and dentine, including the definition of hardness, methods for the determination of hardness and the relation of tooth hardness to such factors as age, sex, type of tooth, location on tooth, caries and fluorosis.

Chapter II considers the effect of variation in plane of

section in the same tooth on hardness values; a variation which has not previously been reported. A comparison of enamel and dentine hardness with some common dental restorative materials is also included.

Chapter III deals with experimental observations on the softening of sound enamel following treatment with lactate buffers. The effect of topical fluoride applications on the rate of softening is also described.

Chapter IV reports some quantitative hardness measurements on teeth from endemic fluoride areas and compares the hardness of these teeth with that of teeth from non-fluoride areas.

CHAPTER I.

THE HARDNESS OF ENAMEL AND DENTINE

REVIEW OF LITERATURE

Definition of hardness

It is impossible to define hardness concisely so as to include all the various characteristics of a material which have been referred to as hardness. The precise definition depends entirely on the method of measurement and no method is dependent on a single physical property. Both elastic and plastic deformation characteristics of a material may be involved, so that the elastic limit, elastic modulus, yield point, tensile strength, brittleness, etc. all play a part in the result obtained (Mott, 1956).

The concept of hardness depends on one's sphere of activity. The housewife thinks of the hardness of materials in terms of the readiness with which they can be cut with a kitchen knife. The fitter judges hardness by the ease with which a material can be sawn, filed or drilled. The dentist refers to hard or soft teeth depending on their resistance to scratching by an explorer or cutting by a bur. The engineer and metallurgist often grade their materials on the basis of penetration of a hard indenter when forced into the surface under controlled conditions. Finally the student considers an examination hard if he has difficulty penetrating the answer.

As early as 1822 Mohs, a mineralogist, wrote "Hardness in general may be defined to be the resistance of solid minerals to the displacement of their particles". Mohs

considered one body harder than another if as a flat surface, it was not scratched by the other body in the shape of a point. He devised a scale for the degrees of hardness based upon ten common minerals as given in Table I. The scale has been of great value. Although still used by mineralogists (Hurlbut, 1959), and at one time by dental investigators, it lacks precision.

Brinell (1906) a Swedish engineer, measured hardness by pressing a steel sphere of known diameter into the surface of the specimen to be tested, under a known load for a standard period of time. The hardness is expressed by the quotient between the load (in kilograms) and the area of the indentation (in sq. mm.).

Many subsequent methods have been based on the same concept - that hardness is a relation between the load and the size of an indentation or scratch. Thus, hardness may be described as the resistance of one body to the "attempt" of another body to occupy the same space at the same time. Hodge and McKay (1933) pointed out that hardness was variously measured as the resistance of a material to scratching, indentation, elastic impact, cutting and/or permanent deformation. Some investigators (Wright and Fenske, 1938; Souder and Schoonover, 1943) included resistance to abrasion or wear. However, other factors are involved in all these properties. Abrasion resistance may not necessarily be proportional to

resistance to indentation. Williams (1942) discussing indentation hardness stated: "The resistance to penetration rests upon the difficulty of pushing the granules composing a body to one side and getting them out of the way of the penetrator. Hardness seems to rest upon the inter-atomic and inter-molecular forces which hold these particles together in a solid. So far as we know, these are common forces and the overcoming of them is a measure of hardness". A workable definition of the hardness of enamel or dentine then is that hardness is a measure of the resistance to permanent deformation or damage.

Two additional terms, microhardness and microindentation have been frequently used and require definition. Microhardness tests usually refer to any technique for determining hardness at low loads, that is, loads of less than 1 kg. Microindentation hardness testing involves a method whereby small dimension impressions are obtained.

Description and comparison of methods for the determination of hardness.

Three distinct types of methods for the determination of hardness have found application in the dental field. These types are: measurements of resistance to penetration (or scratching), abrasion, and elastic deformation.

A comparison of several methods is given in Table I.

The basis of Table I is the arbitrary Mohs scale, based on the ability of substances with high Mohs numbers to scratch substances having low Mohs numbers. The hardness intervals represented by one unit of the Mohs scale are quite variable. Thus, a difference of one unit on the Mohs scale, between calcite and fluorite is equivalent to 28 Knoop units, whereas a difference of one Mohs unit between fluorite and fluorapatite is equivalent to 273 Knoop units. Mohs realized that the intervals between members of the scale were not of the same magnitude. He developed the use of the file (i.e. abrasion test) for subdividing the intervals of hardness on the Mohs scale.

A more refined modification of the scratch principle was Pickerill's sclerometer, (1912) which consisted of a diamond point and weight balanced on a brass rod. The tooth was drawn under the point with gradually increasing pressure until a scratch was made which could be seen macroscopically when rubbed with graphite.

Later scratch methods for studying the hardness of teeth used microscopic examination to measure the width of the scratch made by an indenter with a constant load; in some methods a constant width of scratch was produced by variable loads. As the width of the scratch is related to the shape of the diamond point as well as the hardness of the test substance, the values obtained by different investigators do

not always agree. A further difficulty of the scratch method arises from splintering of the edges of the grooves. The edges of the scratch may be burred due to piling up of material and the correct width cannot be readily measured. The Bierbaum microcharacter hardness tester is a scratch instrument applied to dental tissues by Hodge (1936),

The Herbert Pendulum Hardness Tester, used extensively by Karlström (1927, 1931) for hardness measurement of teeth, consisted essentially of an inverted compound pendulum supported on a steel ball or on a diamond or ruby cone. The ball or cone rested on the test substance and the pendulum was then set rocking in a vertical plane. The time of oscillation (in seconds) required for a given number of pendulum swings was recorded as the time-hardness number. The hardness of the material affects the periodicity, and the decrement of the amplitude of the swing will be less the harder the specimen under test.

The Shore Scleroscope Hardness Tester (Williams, 1942) is partially based on the elastic properties of materials. A tiny diamond-tipped hammer was dropped on a surface and the height of rebound was measured. The hardness was the resistance to penetration as measured by the extent of rebound.

In the dental field, at least, instruments based on measurements of resistance to penetration have replaced those based on resistance to scratching. The usual Brinell and

Rockwell instruments widely used for metals, require larger specimens than can be readily obtained with teeth. The principle of the method is to press a ball under known load into the specimen. The diameter of the impression, related to the load, gives the hardness. The Monotron Hardness Tester measures the load required to give a constant size impression with a ball or diamond cone. Head (1917) designed the Microdynamometer which is a semi-microhardness tester making an indentation of constant size. He measured the depth of the indentation when a known pressure was applied. Head used three punches for the indenter : a heavy steel punch, an iridium-pointed punch and a diamond pointed punch, each 0.02 inch in diameter.

The original method of Brinell has certain limitations. Thus, when the hardness of the test material approached that of the steel sphere, deformation of the indenter as well as the test material occurred. Shattering and breakage of the specimen are common problems with brittle materials.

In 1939, Knoop, Peters and Emerson introduced certain modifications of the indentation hardness test which overcame many of the previous objections to this technique. The indenter consisted of a diamond crystal rigidly mounted in a metal holder. The diamond was accurately ground to pyramidal shape with the included longitudinal angle $172^{\circ}30'$ and the included transverse angle 130°. The indenter was made a part

of one arm of a balance, and could be loaded with weights varying from 1 g. to 500 g. The impressions that resulted from this type of indenter were rhomboidal or "diamond-shaped" with the long diagonal 7 times the length of the short one, and 30 times the depth of the impression. With this indenter, the major part of any elastic recovery of the impression upon removal of the load takes place transversely rather than along the length of the impression (Robertson and van Meter, 1951). Consequently by taking the measurements of the long diagonal only, the unrecovered projected area can be computed. The Knoop Hardness Number (KHN) is expressed as Kg/mm^2 and is the load required to force the indenter down until one square millimeter of surface is occupied. It is given by the formula:

$$KHN = \frac{L}{A_p} = \frac{L}{l^2 C_p}$$

where:

L = load in kilograms.

A_p = unrecovered projected area of indentation in square millimeters.

l = measured length of diagonal of indentation in millimeters.

C_p = 7.028×10^{-2} for a theoretically perfect indenter.

The Knoop Indenter has several advantages particularly in reference to the Brinell method. The dimensions are

strictly defined and reproducible. The diamond indenter does not distort as readily as a steel sphere. The indentation is small and permits pin-point placement. Very little cracking or shattering occurs if care is taken during application and removal of the load. Since its introduction, the Knoop indenter has been widely used in dental research as indicated in the summary table (Table II).

Sometimes a pyramidal indenter of the Vickers or "Square Base" type is used. It is similar to the Knoop indenter but the diamond is ground to a square base pyramidal form with the included angles between faces of 136°. For a theoretically perfect impression, both diagonals are of equal length. In practice, both diagonals are measured, and the average is used in computing the hardness.

Hodge (1936) made an extensive comparison of the hardness of dental tissues with the Brinell, the Rockwell, and the Monotron instruments. He also used the Shore Scleroscope, Herbert Pendulum and the Bierbaum Microcharacter hardness testers. A series of substances were compared and found to fall in the same sequence of hardness values for all the instruments, but relative hardness varied considerably. For some instruments, Hodge noticed fragmentation or distortion of the enamel and dentine, and in subsequent work used the Bierbaum Microcharacter scratch hardness tester exclusively. Richter (1931) compared the results obtained by using scratch

and indentation (Brinell) methods and considered indentation tests best suited for dental tissues.

Most current studies on enamel or dentine hardness are using an indentation tester of which several are commercially available. Only those instruments most commonly used will be described; these included the Tukon Tester (Wilson Mechanical Instrument Division, American Chain & Cable Company, Inc.), the Kentron Tester (Torsion Balance Company), the Hanemann Apparatus (Zeiss Optical Company), and the Durimet Tester (E. Leitz, Ltd.).

The Tukon tester consists essentially of a beam with a diamond indenter at one end. The beam is pivoted on a knife edge and the load is applied by a carrier arm. After the test the stage carrying the specimen is slid into a position so that the indentation is central in the field of a measuring microscope carrying a filar eyepiece. Interchange of Knoop and Vickers indenters is readily carried out.

The Kentron Tester is similar in some respects to the Tukon but has a modified form of loading in which the rate is variably controlled by means of a small hydraulic dash pot. The beam suspension is a flexure plate which provides relatively friction free movement during indentation and restricts the movement of the indenter to a precise predetermined arc.

The Hanemann Apparatus is a self contained unit with the indenter and loading mechanism incorporated in a specially

designed objective lens, which is used for examination of the specimen and for selection of the area to be tested. The load is applied through two parallel disc springs by racking the focussing unit. The size of the indentation is measured by an eye piece micrometer after the instrument has been refocussed to remove the load.

The Durimet Tester consists of a base which carries a table and column on which is fixed the microscope and indenting mechanism. The arm carrying the indenter is lifted by a Bowden cable and at the same time a spring is preloaded. When the spring is released the indenter approaches the specimen at a speed controlled by an oil brake. The spring pressure is automatically cut off when the full load is applied. The indentation is measured by means of one or two objectives which are mounted together with the indenter on a rotating head.

Hardness of enamel, dentine and related mineral crystals

Table II summarizes in chronological sequence the values reported by various investigators on the hardness of enamel and dentine. Many different techniques have been used and consequently the findings have been expressed in different units. When possible, the estimated equivalent in Knoop Hardness units is given in parentheses. These estimated

values probably should be considered only as gross approximations.

A wide range of hardness values have been reported for enamel. Wedl (1872) and Kuhns (1895) reported extremely high values, equivalent to 820 KHN and 560-1340 KHN, respectively. Neither of these authors reported the method used in measuring the hardness, and in view of later findings it is doubtful if their values are accurate. Most investigators (Karlström, 1927; Paffenbarger et al., 1938; Richardson and Worner, 1945; Swartz and Phillips, 1952; Bhussry, 1958; and Skinner, 1957) have found hardness values for enamel in the range of 160-300 KHN. These studies usually were for ground enamel surfaces or tooth sections rather than intact surface enamel. Recent studies (Caldwell et al., 1957-1958; Craig and Peyton, 1958; Newbrun et al., 1959) have reported the hardness of intact and ground enamel surfaces as falling in the range of 300-400 KHN.

Dentine is much softer. The hardnesses reported do not differ as much as for enamel and fall in the range of 60-150 KHN, mostly 60-80.

Enamel compares in hardness with certain of its related minerals. It is apparently somewhat softer than fluorapatite, (430 KHN) but is considerably harder than fluorite (163 KHN). Values for hydroxyapatite are not available.

Variation in the hardness of mineral crystals has been ascribed to changes in the composition, to changes produced

by variously oriented crystal grains in the aggregate, and to changes in directions in which the hardness test was made with respect to the crystal faces and planes of cleavage of the crystal as a whole. Although the individual crystals in enamel are very minute, it is probable that the plane in which the hardness measurement is made still will have some effect on the hardness. Different investigators have used frontal, sagittal or coronal sections of teeth or the intact surface. Such differences may account in part for the wide variation of values reported.

Relations of tooth hardness to other factors.

Effect of age.

Proell and Schubert (1928), Karlström (1931) and Richter (1931) claimed that the enamel of deciduous teeth was softer than that of the permanent dentition. Wright and Fenske (1938) also found variation of hardness with age, maximum hardness occurring between 31-40 years. Atkinson and Saunsbury (1953) stated that "mature" deciduous teeth (those naturally shed) were hardest of all and that permanent teeth soften with age. Nihei (1959) claimed that enamel, dentinal and cemental hardness increased with age. Dalitz (1961) failed to show any relationship between dentinal hardness and age. Caldwell et al. (1957) found no pronounced hardness difference between deciduous, permanent, erupted and unerupted

teeth nor between teeth from individuals of different ages. No clear evidence exists for an effect of age.

Sex factors.

Three investigations have been made of variation in hardness depending on sex. Kuhns (1895) found the enamel of females to be harder than that of males, and Wright and Fenske (1938) found the dentine of females to be significantly harder than that of males. Proell and Schubert (1928) observed no sex difference in the hardness of teeth. Further work on tooth hardness in relation to sex is necessary before any conclusions are justified.

Type of tooth.

Caldwell et al. (1957) compared the hardness of the enamel surface of different tooth types as shown in Fig. I. They could find no significant difference between the hardness of human anteriors (365 ± 35 KHN) and posteriors (393 ± 50 KHN). Bremer (1938) studying the hardness of monkey and dog teeth noted no difference between the hardness on the right or left side, nor between nervated and denervated sides. Cows' teeth are of similar hardness to human teeth (Newbrun et al. 1959).

Location on tooth.

Most reports, (Schultz, 1898; Burg, 1921; Richter, 1931; Karlström, 1931; Hodge and McKay, 1933; Hodge, 1936; Atkinson

and Saunsbury, 1953; Caldwell et al., 1958; Newbrun et al., 1959; Nihei, 1959) of variation in the hardness of enamel at different anatomic locations of tooth sections state that enamel increases in hardness from the DE junction towards the surface or that the surface enamel is harder than the subsurface enamel. However, Gustafson and Kling (1948) and Craig and Peyton (1958) could observe no definite trend in hardness of enamel from the DE junction to the outer surface. Gustafson and Kling suggested that, as there are many soft tufts near the DE junction, these tufts were the cause of the low values at the junction; they concluded that the enamel has a constant hardness throughout its depth.

Intact surface enamel is harder than ground and polished subsurface enamel (Caldwell et al., 1958; Newbrun et al., 1959). However, this difference can not be accounted for by the proposal of Gustafson and Kling because there are no tufts immediately below the surface. Possibly, the direction or plane in which the measurements were made may have had some effect on the hardnesses.

Karlström (1931) claimed that the hardness of ground enamel on the buccal surface increased from the cervical region towards the incisal edge. Enamel decreases in thickness towards the cervical region. As Karlström ground the teeth to prepare plane surfaces he was closest to the DE

junction in the cervical region. His data were also based on only four hardness determinations at the cervical region and are of doubtful validity. Subsequent studies have failed to indicate any such trend.

Most workers agree that extremely local variations in enamel hardness exist even in adjacent areas of enamel in a single tooth (Swartz and Phillips, 1952; Atkinson and Saunsbury, 1953; Caldwell et al., 1957; Craig and Peyton, 1958). In Fig 2, two adjacent impressions of the Knoop indenter show such local variations.

From studies on dentine it has been variously concluded that the dentine adjacent to the pulp is softest (Bremer, 1938; Craig et al., 1959), that root dentine is softer than coronal dentine (Hodge and McKay, 1933) and that the hardness of root and crown dentine is the same (Craig et al., 1959).

Carious teeth.

As one would expect, naturally carious areas have been shown to be softer than sound enamel (Bhussry, 1956, 1958; Newbrun et al., 1959; Sognnaes, 1959). Some investigations have claimed that sound enamel which was "sclerotic" (hard) is more resistent to caries than sound enamel which is "malacotic" (soft). But both clinical and experimental research suggest that the hardness of sound enamel is independend of the amount of caries (Hodge, 1939; Nihei, 1959), of the

enamel solubility (Thurlow and Buntzell, 1927; Swartz and Phillips, 1952), and of the rate of enamel softening in acid buffer (Newbrun et al., 1959).

Hardness appears to be a sensitive and suitable criterion for assessing the progress or rate of naturally and artificially carious lesions. Leber and Rottenstein (1867), Miller (1883) and Mummery (1910), as well as many of the more recent workers (Dobbs, 1932; Hoff and Köszeg, 1937; Swartz and Phillips, 1952; Caldwell et al., 1958; Newbrun et al., 1959; Sognnaes, 1959) have successfully used hardness as a measurement of decay. Hardness measurements of naturally carious lesions cut in section (Gustafson, 1959; Sognnaes, 1959) have confirmed results obtained by radiographic and polarized light examination. The surface layer was harder than the underlying parts of the lesion and carious enamel was softest at the subsurface zone. However, no evidence exists that teeth prone to caries are initially soft.

Hodge (1937) and Craig et al. (1959) drew attention to changes in the hardness of dentine occurring as a result of caries. Carious dentine was much softer than normal dentine, but immediately surrounding the soft carious dentine was a narrow zone of dentine which was of greater hardness than normal dentine. They differed in their findings concerning the hardness of transparent dentine which Hodge found less

than normal, and Craig et al. found slightly greater than normal.

Sound dentine, in non-carious teeth may also change in hardness following chemico-mechanical intervention. Mjør et al. (1960) have reported a significant increase in hardness of calcium hydroxide covered dentine as compared with unaffected dentine in the same tooth, dentine in unoperated teeth and dentine under amalgam restorations. The hardness changes occurring in dentine appear to be a result of pulpal stimulation caused by caries or calcium hydroxide whereas amalgam does not produce sufficient stimulation to alter the hardness of the underlying dentine.

Effect of fluorides on hardness of enamel.

One of the earliest references to the effect of fluoride on enamel hardness was contained in a paper by Bowes and Murray (1936) who noted on grinding fluorosed enamel that it seemed less hard than normal. Phillips and Swartz (1952) used topical applications of fluoride to study the effect on the hardness of ground enamel. Following a five minute immersion in acidified sodium fluoride solution, they claimed a 5.1% increase in enamel hardness, and with stannous fluoride a 7.1% increase. Similarly, Hord and Ellis (1949) using dogs found a 13% increase in enamel hardness after *in vivo* topical applications of sodium fluoride. Neither of

these studies gave any statistical data. In consideration of the inherent wide variation in hardness of enamel, these slight increases seem insignificant.

More recently Caldwell et al. (1958) and Newbrun et al. (1959) found that following topical application of fluoride, no demonstrable change in hardness could be determined greater than the error of testing. Both studies showed that topical fluorides could reduce the rate of softening of ground enamel surfaces, but not of intact surfaces. Herrmann (1958) and Herrmann and Rozeik (1959) fed albino rats a diet containing 500 or 1,000 ppm F and reported both topical and systemic effects on tooth hardness. An increase in hardness was observed for rat molars already formed, whereas there was a decrease for the incisor teeth which were still forming. They tested both the enamel and the dentine and averaged these readings for each tooth.

Newbrun (1960), in an investigation using human teeth from areas having different fluoride concentrations in the drinking water, ranging from 0.00 to 5.0 ppm, observed an inverse relationship between the enamel hardness and the degree of mottling. The enamel of teeth from very high fluoride areas was significantly softer ($P \leqslant 0.01$) than enamel of teeth from an area with no fluoride. Teeth from areas with optimal amounts of fluoride did not differ significantly

in hardness from control teeth.

Rehardening of softened enamel

Considerable emphasis has been placed on the softening that occurs in enamel in both natural and artificial caries but the possibility of rehardening has been largely ignored. As early as 1917, Head reported that teeth which had been softened so that they could be "readily pared with a lancet", could be rehardened by storing in saliva for several weeks. Further work using the Microdynamometer to test changes in hardness of enamel surfaces, convinced him that saliva could reharden enamel which had been softened by acids.

Recently, Koulourides and Pigman (1960 a,b.) found that artificially softened enamel can be rehardened in solutions of secondary and tertiary calcium phosphate adjusted to pH 6.8 to 7.3 and maintained at 37°C. Rehardening took place over a period of one to two weeks, the solutions being changed daily. With some teeth, the final hardness was greater than the initial hardness, the percentage recovery of hardness varying between 42-128%. Tricalcium phosphate gave a faster recovery than did dicalcium phosphate and the rehardening was further accelerated by the addition of fluoride. The recovery of hardness was not complete when enamel was softened below 170 KHN. Further studies are necessary to determine whether these *in vitro* rehardening procedures have any clinical applications.

Table I.

Comparison of hardness values of various materials on Mohs (1822), Beirbaum Microcharacter (Hodge and McKay, 1934), Knoop (Foster, 1956) and Brinell Scales (Wilson Chart 52)

Substance	Formula	Mohs Scale	Bierbaum Micro-character	Knoop Hard-ness Number	Brinell Hardness Number
Talc	$3MgO.4SiO_2.H_2O$	1	1	-	-
Gypsum	$CaSO_4.2H_2O$	2	11	32	-
Calcite	$CaCO_3$	3	129	135	107
Fluorite	CaF_2	4	143	163	130
Fluorapatite	$CaF_2.3Ca_3(PO_4)_2$	5	577	430	400
Feldspar	$K_2OAl_2O_3.6SiO_2$	6	975	560	500
Quartz	SiO_2	7	2700	820	-
Topaz	$(AlF)_2SiO_4$	8	3420	1340	-
Corundum	Al_2O_3	9	5300	2100	-
Diamond	C	10	-	7000	-

TABLE II.

Compilation of enamel and dentine hardness values obtained by different investigators*

Author(s) Year	Hardness (KHN = Knoop Hardness)		Method employed	Conclusions
	Enamel	Dentine		
Hoppe 1862	-	-	Scratch resistance, relating enamel to other minerals	Enamel hardness similar to apatite. No difference in hardness of different layers.
Kopesky 1870	-	5-6 Mohs (430-560 KHN)	Not described	-
Wedl 1872	7 Mohs (820 KHN)	-	Not described	-
Kuhns 1895	5-8 Mohs (560-1340 KHN)	-	Not described	Male teeth slightly softer (5-6 Mohs) than female teeth (7-8 Mohs).
Schultz 1898	-	-	Not described	Enamel has same hardness as apatite.

*The hardness values reported by the original investigators are given in the numbers without parentheses. The Knoop Hardness values (KHN) given in parentheses are estimated from the relation between the Knoop scale and the other scales shown in Table I.

Author(s)	Hardness (KHN = Knoop Hardness)		Method employed	Conclusions
	Enamel	Dentine		
Pickerill 1912	-	-	Scratch Sclerometer	"Sclerotic" teeth hard and caries immune. "Malacotic" teeth soft and caries susceptible
Head 1917	-	-	Microdynamometer	Saliva can reharden soft enamel. Enamel surface harder than deeper layers.
Burg 1921	-	-	Scratch	Hardness of enamel varies with direction of scratch. Dentinal hardness independent of direction. Outside surface of enamel gave highest value.
Karlström 1927, 1931	20.9 (4Mohs Scale or 163 KHN)	13 (3-4 Mohs or 135-163 KHN)	Herbert Pendulum Tester measuring time for 5 swings. Used ground surfaces	Hardness of enamel on buccal surface increased from cervical towards incisal edge. Enamel hardest in external layer and drops rapidly as DEJ is approached. Deciduous teeth have slightly lower hardness than permanent teeth. No change in hardness following storage in 5% formalin

Author(s)	Hardness (KHN = Knoop Hardness) Enamel	Dentine	Method employed	Conclusions
Proell & Schubert 1928	-	-	Scratch on teeth out in sagittal section	No sex difference in hardness of teeth. Deciduous teeth softest. Hardness increases with age. Hardness in sagittal section increases towards surface.
Richter 1931	-	-	Scratch on sagittal sections similar to above. Also indentation (Brinell) method.	Enamel towards surface hardest; dentine adjacent to pulp softest. Increase in hardness after drying. Deciduous teeth much softer than permanent.
Hodge et al. 1933-39	300-1000 av.600 (440 KHN)	121 (135 KHN)	Bierbaum microcharacter scratch method on sectioned teeth	Outer shell of enamel had maximum hardness with progressive softening towards DEJ. Root dentine softer than coronal dentine.
	22-27	12	Herbert Pendulum Tester time/5 swings	Transparent secondary dentine softer than normal. Hardness was independent of the amount of caries in mouth.
Wright & Fenske 1938	-	-	Extent of abrasion under constant load	Male dentine significantly softer than female dentine. Hardness varies with age; maximum hardness between 31-40 years.

Author(s) Year	Hardness (KHN = Knoop Hardness) Enamel	Dentine	Method employed	Conclusions
Paffenbarger et al. 1938	232-322 KHN	50-60 KHN	Knoop hardness tested on transverse sections	Marked difference in hardness of enamel and dentine
Bremer 1938	-	12-13	Herbert Pendulum Tester measuring time/5 swings; teeth of dogs and monkeys	Denervation does not affect hardness. Dentine decreases in hardness toward the pulp, is softest in younger teeth. High degree of variation of enamel hardness even in adjacent points and especially at DEJ.
Klinger 1940	140-400 BHN (154-430 KHN)	70-120 BHN (91-135 KHN)	Brinell Hardness Number using a Vickers tester on ground surfaces.	Caries-free teeth harder, 301 BHN (330 KHN) than carious teeth, 217 BHN (231 KHN). Extreme variation in enamel hardness. No relation between hardness and solubility.
Totah 1942	-	-	Bierbaum microcharacter-scratch method	Dentine increased in hardness following drying

Author(s) Year	Hardness (KHN = Knoop Hardness) Enamel	Dentine	Method employed	Conclusions
Hinds 1943	–	–	–	No difference in enamel hardness of erupted and unerupted teeth. No correlation between age and hardness of permanent and deciduous teeth
Souder & Schoonover 1943	350 KHN	50 KHN	Knoop indenter	Hardness good method of following in vitro caries
Richardson & Worner 1945	255 KHN	60 KHN	Knoop diamond in which the load was applied by a wound spring	–
Gustafson & Kling 1945, 1948	310 DPH (318 KHN)	–	Hanemann tester with diamond pyramid indenter; 9-30 g. load on sections of teeth. Results expressed as Diamond Pyramid Hardness (Vickers)	No difference in hardness between freshly extracted teeth and those allowed to dry out. Tufts and spindles were softer than surrounding enamel. Striae of Retzius usually of normal hardness; in isolated cases harder than adjacent enamel. Enamel hardness approx. same throughout thickness; softest near DEJ due to tufts

when cavitation was evident. Such areas had hardness values usually falling in the range 100 to 250 KHN. This range corresponds to that for ground enamel surfaces after exposure to the buffer for about 8 to 10 hours.

Integrity of enamel surface during the softening process.

In order to determine whether appreciable loss of surface material occurred during the softening process produced by the buffer, the original indentations were re-measured after the 8-hour period. The lengths of the indentations showed no significant decrease (corresponding to less than 1.5 microns in length or 6 KHN), although the teeth had dropped in hardness to the range of 125 to 200 KHN.

TABLE VII

Average rate of softening of enamel on exposure to lactate buffer

	Time in hours					
	2	3	4	6	8	10
	Intact Enamel (Human)					
KHN drop/hr	19.5	16.0	22.0	23.0	20.0	19.0
% KHN drop/hr	5.3	4.5	6.1	5.7	5.4	5.1
	Ground Enamel (Human)					
KHN drop/hr	22.9	20.2	22.5	22.7	23.1	22.0
% KHN drop/hr	7.3	6.4	7.9	7.8	7.5	6.9
	Intact Enamel (Bovine)					
KHN drop/hr	35.9	-	-	27.6	-	21.8
%KHN drop/hr	10.0	-	-	7.4	-	6.2

TABLE VIII

Average effect of fluoride treatment on the rate of softening of human enamel after 8 hours

Treatment	Initial hardness (KHN ± S.D.)	Hardness after lactate treatment (KHN ± S.D.)	Drop in hardness (KHN)	% drop (% initial KHN)	KHN drop/hr	% KHN drop/hr ± S.D.
		Intact Human Enamel				
None	369 ± 36	207 ± 25	162	43.5	20.0	5.4 ± 1.0
2% NaF	377 ± 36	223 ± 38	154	41.1	19.3	5.1 ± 0.8
		Ground Human Enamel				
None	310 ± 22	124 ± 23	186	59.9	23.1	7.5 ± 0.9
2% NaF	267 ± 28	141 ± 12	126	47.2	15.7	5.8 ± 0.9

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Author(s) Year	Hardness (KHN = Knoop Hardness) Enamel	Dentine	Method employed	Conclusions
Phillips & Swartz 1948	-	-	Tukon tester with Knoop indenter 500 g. load on ground enamel	Fluoride compounds in vitro increased the hardness of enamel although the change was small
Hord & Ellis 1949	-	-	Tukon tester with Knoop indenter	Topical applications of NaF in vivo increased hardness of enamel of dog
Swartz & Phillips 1952	160-230 KHN	-	Tukon tester with Knoop indenter 500 g. load on ground enamel	Enamel varied widely in hardness and solubility. No correlation between hardness and solubility
Atkinson & Saunsbury 1953	-	-	Vickers diamond with 30 g. load applied by air pressure on tooth sectioned bucco-lingually	Wide variations in hardness of enamel across sections of teeth especially deciduous teeth. Mature "deciduous" teeth hardest. Permanent teeth soften with age. Gradual decrease in hardness towards DEJ.
Steel 1955	356 DPH (365 KHN)	-	Vickers hardness tester with 5 Kg. load on intact enamel surfaces	No correlation between hardness and solubility of intact tooth surface

Author(s) Year	Hardness (KHN = Knoop Hardness)		Method employed	Conclusions
	Enamel	Dentine		
Muntz et al. 1956	300-407 KHN	–	Kentron Microhardness tester with Knoop Diamond indenter used on intact enamel surfaces; 500 g. load	See Caldwell et al.
Bhussry 1956, 1958	252 KHN	–	Tukon tester with Knoop indenter 200 g. load on tooth sections	Brown spot areas lowest hardness, 95 KHN. Consolidated zone intermediate hardness, 142 KHN. Sound enamel hardest 252 KHN
Skinner 1957	290 KHN	65-80 KHN	–	Hardness of dentine and silicate cements essentially the same
Gustafson 1957	–	–	Hanemann tester with diamond pyramid indenter. Results expressed as Vickers Hardness number	Microhardness tests on carious lesions confirmed polarized light findings. Surface layer is harder than underlying parts of lesion.

Author(s) Year	Hardness (KHN = Knoop Hardness) Enamel	Hardness (KHN = Knoop Hardness) Dentine	Method used	Conclusions
Caldwell et al. 1957	380 KHN	-	Kentron Microhardness tester with Knoop diamond indenter used on intact enamel surfaces; 500 g. load	Extensive variation in hardness of areas on same tooth with no consistent pattern. No pronounced difference between deciduous, permanent, erupted and unerupted teeth, nor between teeth from individuals of different ages
Caldwell et al. 1958	367 KHN	-	As above	Topical applications of fluoride retarded softening of enamel treated in "artificial" mouth. Subsurface enamel softer than intact surface enamel
Craig & Peyton 1958	343 ± 23 KHN	68 ± 3 KHN	"MO" Tukon tester with Knoop diamond indenter 50 g. load on transverse sections of teeth.	Pronounced difference in hardness values of different sections of the same tooth. Range - 272-400 KHN. No definite trend in hardness of enamel from DEJ to the outer surface, nor from crown to cervical margin.

Author(s) Year	Hardness (KHN = Knoop Hardness) Enamel	Dentine	Methods employed	Conclusions
Craig et al. 1959	-	65-72 KHN	As above, using a 10g. load on dentine	Dentine near DEJ softer than surrounding dentine. Dentine adjacent to pulp chamber 30 Knoop units lower. Hardness of root and crown dentine same. Carious dentine much softer than normal, whereas surrounding dentine slightly harder than normal
Newbrún et al. 1959	365 ± 35 KHN	-	Kentron micro-hardness tester with Knoop diamond indenter and 500 g. load on intact enamel surfaces (anteriors)	Rate of softening of teeth in acid buffer was independent of initial hardness. Fluoride treatment reduced rate of softening of ground surfaces but not intact surfaces. Naturally carious areas had hardness 100-250 KHN
Newbrun 1959, 1960	-	-	As above, using teeth from different areas where F conc. in drinking water ranged from 0.00 - 5.0 ppm	Teeth from very high F areas were significantly softer than teeth from an area with no F. Teeth from areas with optimal amounts of F did not differ significantly from control teeth. The Knoop Hardness appeared to be inversely proportional to degree of dental fluorosis

Author(s) Year	Hardness (KHN = Knoop Hardness) Enamel	Dentine	Method employed	Conclusions
Herrmann et al. 1958, 1959	-	-	Durimet tester using a 50 g. load. Results expressed in microns	For rats fed high fluoride diets, enamel of teeth already formed increased in hardness whereas enamel of teeth still forming was softer than normal
Nihei 1959	310-349 DPH (320-356 KHN)	-	Durimet tester using a 50 g. load measuring Diamond Pyramid Hardness on tooth sections	Enamel hardness decreases from surface to DEJ. Enamel, dentinal and cemental hardness increase with age. Enamel harder at cusp than at cervical. Enamel hardness markedly lower in horizontal than in longitudinal sections. Sound enamel of carious teeth has same hardness as normal enamel. Enamel hardness of mottled teeth lower than normal

Author(s) Year	Hardness (KHN = Knoop Hardness)		Method employed	Conclusions
	Enamel	Dentine		
Sognnaes 1959	317 KHN	75 KHN	Kentron microhardness tester with Knoop indenter on tooth sections	Carious enamel softest at the subsurface, 12 KHN, intermediate hardness at the central zone, 190 KHN and soft again at the inner zone, 139 KHN. Carious dentine 43 KHN at subsurface 52 KHN at central zone and 63 KHN at inner zone
Avery & Visser 1959, 1960	-	-	Tukon tester with Knoop indenter on sections of human and monkey teeth	Followed the mineralization pattern of developing enamel using hardness tests
Koulourides & Pigman 1960	-	-	Kentron microhardness tester with Knoop diamond indenter and 500 g. load on ground surfaces	Artificially softened enamel was rehardened in $CaHPO_4 \cdot 2H_2O$ over a period of 1-2 weeks
Mjør et al. 1960	-	-	Kentron microhardness tester	$Ca(OH)_2$ covered dentine showed a significant increase in hardness compared with unaffected dentine

Author(s) Year	Hardness (KHN = Knoop Hardness)		Method employed	Conclusions
	Enamel	Dentine		
Dalitz 1961	-	52-65 KHN	Durimet tester using a 240 g. load with Knoop indenter on anterior teeth 14-56 age group	No relationship between dentine hardness and age

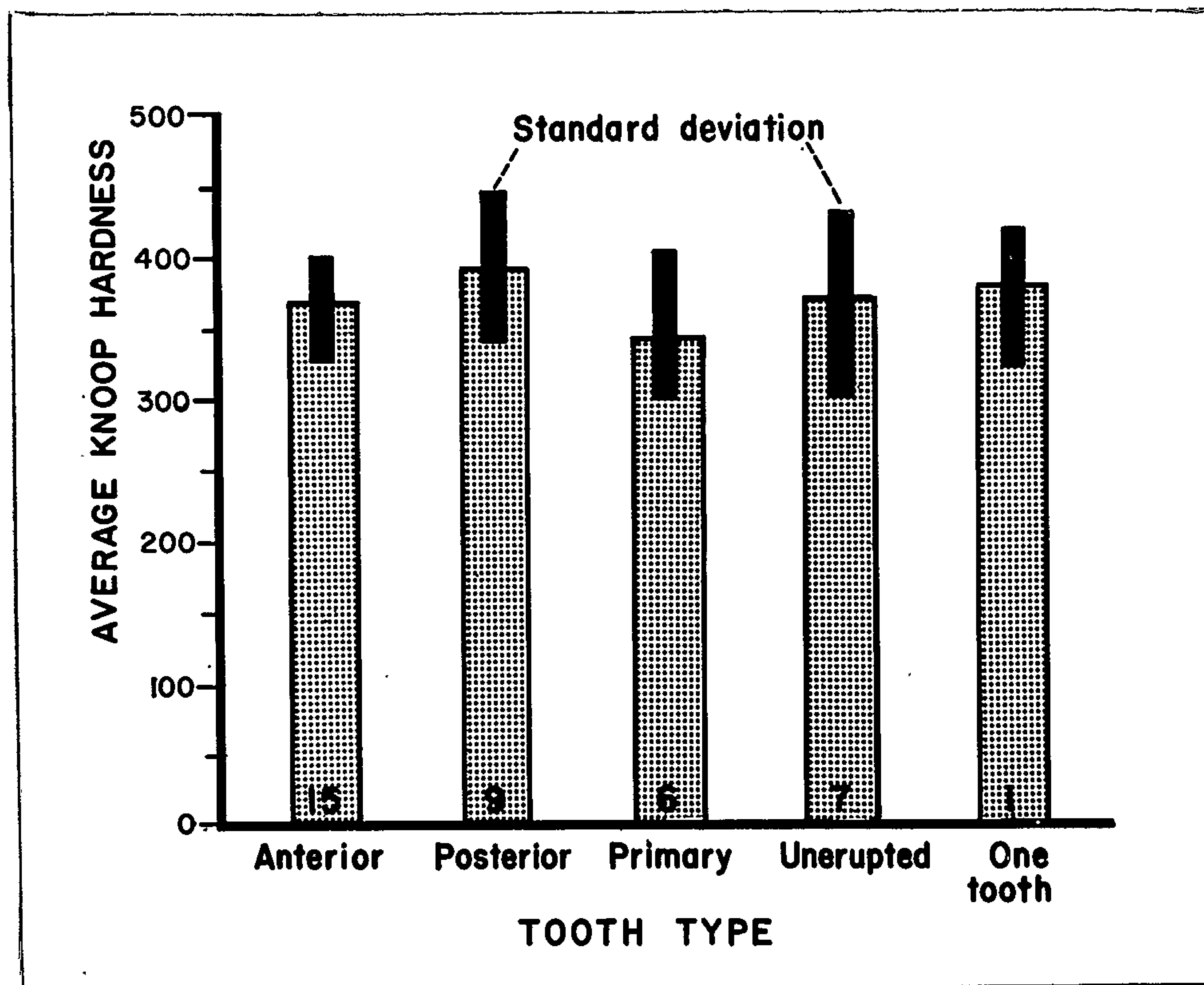

Fig. 1. Comparison of the hardness of intact enamel surfaces of human teeth. (The number at the base of each column indicates the size of the sample.)

Fig. 2. Impressions approximately 0.06 millimetres apart on the external surface of enamel showing local difference in hardness.

CHAPTER II.

THE HARDNESS OF ENAMEL IN VARIOUS PLANES OF SECTION.

The most striking characteristic of enamel is its hardness. The gastric mucosa withstands a markedly acid environment (pH 2-3) which enamel willnot; many organs, epithelia and connective tissues have the property of regeneration which enamel does not have; but in its ability to resist permanent deformation by indentation or by scratching (i.e. hardness) enamel surpasses all other tissues in the body.

The hardness of enamel has been a subject of considerable attention in the dental literature, a wide range of hardness values having been reported. The work described in this chapter was undertaken in an endeavour to find a logical explanation for some of these differences in enamel hardness. Variation in plane of section in the same tooth caused a change in hardness measurements observed. Such an effect has not previously been reported.

The practical significance of the hardness of the enamel is evident. The enamel surface is subject to abrasion, depending on the tooth-brushing habits of the patient (Mannerberg, 1960). In addition to such factors as frequency of tooth-brushing, pressure applied to the tooth-brush, quality of the brush and abrasive ingredients of the dentifrice, the hardness of the enamel surface is an important factor in determining the amount of abrasion which occurs. Attrition, though primarily dependent on

dietary consistency, bruxism or malocclusion, may also be related to the hardness of the enamel. Both clinical and experimental research indicate that the amount of caries (Hodge, 1939), the enamel solubility (Swartz and Phillips, 1952) and the rate of enamel softening in acid buffer (Newbrun et al., 1959) are independent of the hardness of the sound enamel. However, the softening of enamel is the most widely used clinical criterion in the diagnosis of carious lesions. In the operative preparation of teeth the hardness of the enamel and dentine plays an important role, not merely by affecting the instrumentation, but also by helping the clinician determine the extent of the lesion.

MATERIALS AND METHODS

Anterior permanent teeth collected from persons who were residents of Sydney, Australia, were used in this study. The teeth were mounted with the labial surface exposed in an epoxy resin.* Hardness measurements were made on the intact labial surface using a Tuken hardness testing machine with a Knoop indenter and a 500 g. load. The labial surface was then ground on silicon carbide paper Grit Nos. 220, 400 and 600 in that sequence under a water spray. This was followed by grinding on an alumina wax lap and then on a rotating turntable with a diamond paste (4-8 micron particle size and 0-1 micron particle size). Final polishing was effected with a magnesium oxide paste. This produced what will subsequently be referred to as a polished labial surface. Hardness measurements were made on this surface after which the tooth was sectioned longitudinally in a bucco-lingual plane. One of the halves was next sectioned in a transverse plane. The freshly cut surfaces were polished as above and their hardness determined. Filar measurements were converted to the Knoop scale using the special purpose slide rule designed by Samuels and Mulhearn (1953). The mean of ten impressions was taken as the hardness value of each surface and the standard deviation calculated. The results are quoted as Knoop Hardness Numbers plus or minus the

*Epirez Compound No. 2659 E (Epimount).

standard deviation.

Several of the sections were subsequently stained by a modification of Gömöri's silver impregnation of reticulum to bring out morphological detail.

RESULTS

Hardness of the intact surface

The surface hardness of six teeth was measured and fell within the range of values reported previously (Newbrun, 1960) for the surface hardness of teeth from areas of comparable fluoride concentration (Table III). It is interesting to note that the previous values were obtained with a different type of tester but the same load.

Effect of orientation of specimen on Knoop Hardness Number

The polished labial surfaces of the teeth were tested by placing the indentations parallel to the long axis. The specimens were then rotated through 45° and 90° and a further ten measurements taken in each case. The indentations were placed as near as possible to limit any regional differences. In one tooth, the hardness ranged from 338 ± 9 KHN at right angles to the long axis to 329 ± 6 KHN parallel to the long axis and in the other tooth the values ranged from 342 ± 9 KHN to 300 ± 12 KHN (Figure 3.).

Effect of location and plane of section on enamel hardness

When the teeth were sectioned in different planes and hardness determined, some interesting differences became apparent (Table IV). Intact surfaces gave higher values than polished surfaces irrespective of the plane of section. In a transverse plane the location of the indentation had an

important bearing on the hardness value. In all four teeth examined the hardness values were significantly greater near the surface than near the dentino-enamel junction. The variation of hardness in tests near the surface and near the dentino-enamel junction on three teeth cut in a longitudinal bucco-lingual plane does not permit any definite conclusion.

When the values for hardness in different planes of section were compared, it was found that the hardness near the surface of enamel cut in a transverse plane was greater than the hardness in corresponding location cut in a longitudinal bucco-lingual plane; this difference was highly significant ($P < 0.01$). On the other hand, near the dentino-enamel junction no such significant difference in hardness was found in the two planes.

An examination of sections showed that different planes of section influenced the degree of fracturing caused by the indenter. In comparing Figures 3, 5 and 6 of sections cut in different planes, the indentations in Figure 3 showed no evidence of fracturing around the margins. Fracturing around the indentations in a transverse plane was slight (Figure 5) but quite pronounced in longitudinal bucco-lingual plane (Figure 6).

When the enamel was stained to reveal the enamel prisms and interprismatic material (Figures, 4,7,8 and 9), the wavy

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When the enamel was stained to reveal the enamel prisms and interprismatic material (Figures, 4,7,8 and 9), the wavy

course and oblique direction of the enamel rods could be observed in some planes. Near the surface of the enamel in transverse plane the indentations were meeting the enamel prisms obliquely (Figure 7). In the same plane, and on the same tooth near the dentino-enamel junction the indentations were both "side on" and oblique to the enamel prisms because of their irregular and varied path (Figure 8). The direction of the enamel rods in this plane of section changes as they approach the surface. When cut in longitudinal bucco-lingual section the indentations were mainly "side on" to the enamel rods, but the changing pattern as evidenced by the Hunter-Schreger bands made any generalization difficult.

Tests on dentine, on the other hand (Table V) could not demonstrate any pronounced change in hardness values in different planes of section.

<u>Comparison of enamel and dentine hardness with that of dental restorative materials.</u>

The results of this comparison are summarized in Table VI from which it can be seen that only fused porcelain and the cobalt-chromium alloys approach the hardness range of the enamel surface. A specimen of acrylic resin for restorative purposes gave by far the lowest hardness value of approximately 17 of all the materials tested. This value is about the same as that of denture base acrylic resins.

Although an absolute comparison of hardness between such dissimilar materials is not strictly valid, it does give some measure of the differences in hardness.

TABLE III. Average hardness of the intact enamel surface

Number of teeth tested	Average Knoop hardness ± SD	F concentration in water supply (ppm)	Area
6*	368 ± 20	0.05	Sydney N.S.W.
40**	367 ± 35	0.00 - 0.03	Birmingham Alabama
8**	374 ± 30	0.05 - 0.15	Binghamton New York
10**	369 ± 30	0.05 - 0.1	Schenectedy New York

*using Tukon tester

**using Kentron tester

TABLE IV. Effect of plane of section and location on enamel hardness

Tooth number	Labial surface		Near surface		Near DEJunction	
	intact	polished	transverse section	long. sect. bucco-lingual	transverse section	long. sect. bucco-lingual
10	363 ± 42	326 ± 13	324 ± 19	283 ± 13	284 ± 8	268 ± 15
12	392 ± 29	289 ± 26	328 ± 15	268 ± 14	272 ± 22	252 ± 20
G1	-	338 ± 9	314 ± 11	-	287 ± 11	-
G2	-	342 ± 9	317 ± 23	285 ± 16	270 ± 11	280 ± 22

TABLE V. Dentine hardness in different planes

Tooth number	Plane of section	KHN
10	Transverse	50 ± 2
10	Longitudinal bucco-lingual	62 ± 4
12	Transverse	64 ± 5
12	Longitudinal bucco-lingual	59 ± 5
G1	Transverse	73 ± 4
G2	Transverse	61 ± 4
G3	Longitudinal bucco-lingual	60 ± 3
	Mean	63 ± 5

TABLE VI. Comparative hardness of tooth restorative materials.

Material	Knoop Hardness Number		
	Skinner (1957)	Souder & Paffenbarger (1942)	Author
Enamel	260	267	368 ± 20
Dentine	65	55	63 ± 5
Silicate cement	70	54	-
Zinc phosphate cement	-	36	48 ± 5
Amalgam	90	90	100 ± 5
Acrylic resin	16	20	17 ± 1
Pure gold	32	-	-
Gold foil	-	-	81 ± 7
24K gold(cast)	-	22	-
Gold Type A (soft)	55	-	84 ± 5
Gold Type B (medium)	-	-	108 ± 3
Gold Type C (hard)	-	-	123 ± 7
Porcelain (fused)	-	415	446 ± 22
Cobalt-chromium alloy A	274	-	-
" " " B	344	-	-
" " " C	336	-	-
" " " D	388	-	-
" " " O	-	-	376 ± 46
" " " R	-	-	412 ± 19

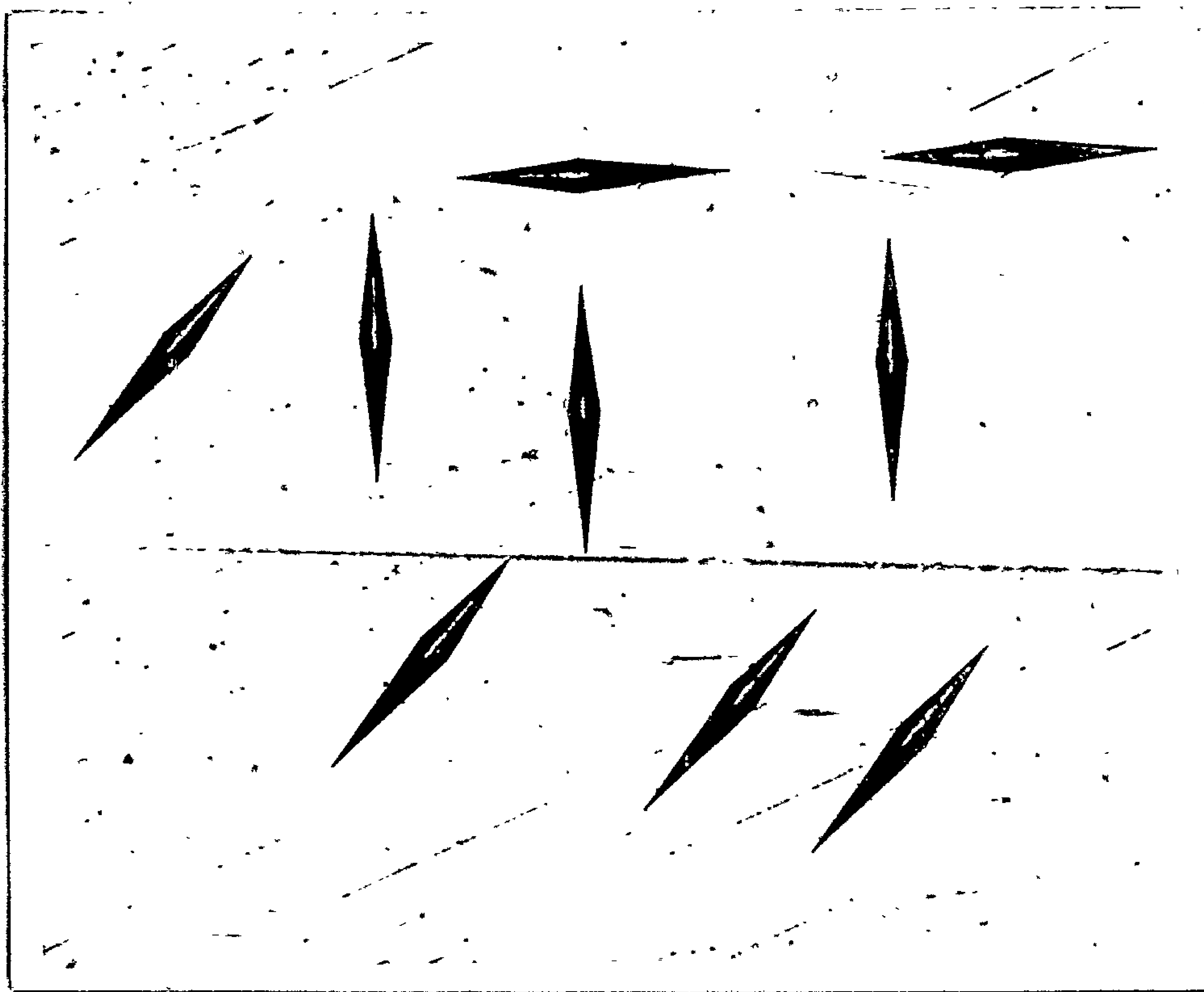

Figure 3. Polished labial surface. Specimen rotated at various angles to Knoop indenter. x 200

Figure 4. Polished labial surface. Gömöri's Silver impregnation stain. Knoop indentations meeting the enamel prisms "end on". x 185 approx.

Figure 5. Transverse section of tooth. Indentations near dentino-enamel junction. x 250

Figure 6. Longitudinal section of tooth in bucco-lingual plane. Indentations near dentino-enamel junction. Fracturing around the indentations is far more pronounced than in the previous figure.
x 250.

Figure 7. Transverse section of tooth. Gömöri's silver impregnation stain. Indentations near enamel surface meeting enamel prisms obliquely. x 185 approx.

Figure 8. Transverse section of same tooth as previous figure. Gömöri's silver impregnation stain. Indentations near dentino-enamel junction. Prisms running a more wavy course in this location. x 185 approx.

Figure 9. Longitudinal section of tooth in bucco-lingual plane. Gömöri's silver impregnation stain. Indentations meeting enamel prisms mainly "side on". Different directions of prism bundles and Hunter Schreger bands readily apparent. Dentine in lower right field.
x 185 approx.

DISCUSSION

From the relevant literature (Table II) it is evident that not only is there a wide variation in the hardness values reported for dentine and enamel, but also in the method employed in measuring this property. Hodge (1936) made an extensive comparison of the hardness of dental tissues with Brinell, Rockwell and Monotron instruments. He also used the Shore scleroscope, Herbert pendulum and Bierbaum microcharacter hardness testers. A series of substances were compared and found to fall in the same sequence of hardness values for all the instruments, but the relative hardness varied considerably. The value for surface hardness 368 $\pm$ 20 in the present study using a Wilson Tukon instrument with a Knoop indenter and a 500 g. load compared exactly with our previous values, (Table III) using a Kentron tester with a similar indenter and load.

Caldwell and co-workers (1957) who had used the same Kentron hardness tester with a 500 g. load reported a mean hardness of 365 $\pm$ 35 KHN for the intact enamel surface of human anterior teeth and 393 $\pm$ 50 KHN for human posterior teeth. They concluded there was no significant difference in the hardness of these two types of teeth. The only other report of the hardness of intact surface enamel was given by Steel (1955) using a Vickers hardness tester with a

diamond pyramid indenter and a 5 Kg. load. A mean of 356 Vickers Hardness Number was found for intact enamel of 148 teeth. This corresponds to about 363 KHN using the Wilson relationship, Table 52 (Wilson Mechanical Instrument Division, American Chain and Cable Company, Inc. N.Y.). The variation between these values and those of most other investigators using similar types of testers could therefore not be attributed solely to differences in instruments.

According to Foley et al. (1960) enamel hardness was load-dependent when tested at low loads. Straight load-dependence usually appears as an increase in hardness with low loads but, if vibration occurs in the testing system, the hardness may decrease with decreasing loads. Only when loads of 10 g. or less are used does this present a problem and, as nearly all studies on enamel hardness have used loads of 50, 200 or 500 g. or more, it is doubtful if the hardness variation could be explained by load-dependence.

Basically, hardness is related to the plastic properties of a material which, in a crystalline substance, varies in different directions in the crystal, i.e. it is plastically anisotropic. Thus, in a single crystal, hardness may be a function of the orientation of the surface in

which indentations are made and also of the directions of the diagonals of the indenter in the plane of the test surface. A pronounced change in hardness value can be produced by changing the orientation of the indenter relative to the surface. Mott and Ford (1954) have shown a 400% variation, using a Knoop indenter on cold rolled magnesium sheet. Moreover, similar variation may be found in a polycrystalline material which has a strongly developed preferred orientation (Crow and Hinsley, 1946).

Enamel is a plastically anisotropic material and these results did show some variation of hardness by rotating the indenter to the specimen in one particular plane. The apatite crystals of enamel are extremely small, their length having been variously estimated as from .027 to 5 microns (Scott and Nylen, 1960). The average size of a hardness indentation under the present conditions was approximately 140 microns which meant that it was not only many times larger than any apatite crystal but also that it was imposed on several enamel prisms (average diameter 4 microns) at the one time. On a polished labial surface the indenter met the prisms "end on" irrespective of the rotation of the indenter (Figure 4). It is not necessary for the indenter to impinge on a single crystal to observe directional hardness differences; if preferred orientation exists the size of the impression is immaterial.

The complex structure of human dental enamel has long been recognized, and has been considered in detail elsewhere (Gustafson, 1945; Orban, 1953). In various planes of section hardness differences were evident (Table IV), and may be accounted for by architectural variation of the enamel prisms and organic matrix as shown in Figures 4, 7, 8 and 9. One of the earliest indications of directional hardness properties was obtained from scratch hardness tests on minerals. Proell and Schubert (1928) attributed differences in the width of scratches on enamel to varying lines of cleavage and to different directions of enamel prisms. However, no experimental evidence was offered to justify this observation. More recently, Nihei (1959) found the hardness of enamel was markedly lower in horizontal (transverse) sections than in longitudinal sections cut bucco-lingually. His results are in direct contrast to the present observations which indicated a lower hardness in longitudinal bucco-lingual section than in transverse section of the same teeth. Near the dentino-enamel junction the hardness difference in the two planes is far less pronounced as the prisms in this location in transverse section are both oblique and "side on" to the indentation. Near the surface, however, the difference is quite marked.

It is suggested that many investigators in the past who have quoted Knoop hardness values for enamel in the 200

to 300 range were testing sections in a longitudinal bucco-lingual plane. In view of the previous discussion it is not surprising that their values were lower than those reported for the enamel surface. Microradiographic studies (Applebaum, 1940; Thewlis, 1940; Newbrun et al., 1959; Soni and Brudevold, 1959) have shown the surface layer of enamel to be more densely mineralized and this would provide an additional explanation of the higher surface hardness. Craig and Peyton (1958) used transverse sections of enamel, and interestingly enough their value of 343 $\pm$ 23 was intermediate between those of other workers presumably made on longitudinal bucco-lingual sections, and our values of 360-380 on the surface enamel.

By tabulating the Knoop hardness number of some commonly used tooth restorative materials one can relate such materials to the enamel hardness. It is not suggested that hardness be the single or the most important criterion in selecting such a material; obviously it is not. The clinician must make his choice, treating each case as an individual problem but perhaps Table VI may help him to reach a decision.

The change in hardness observed with different directions of the enamel rods also has a practical application. Hand instruments will more readily cut enamel along the direction of the rods than across them because

the hardness is less and the rods will cleave along that plane.

SUMMARY

The same hardness results were observed when tested by two different types of instruments using a similar shaped indenter and the same load. The hardness values were significantly different when tests were made in different planes of section and in different locations in the enamel. An explanation based on the architectural arrangement of the enamel prisms and their matrix has been offered for these variations in hardness. Many previous tests quoting a low hardness for enamel appear to have been made in a plane where fracturing and cleavage along the prisms is greatest. The hardness of some commonly used dental restorative materials has been compared with that of enamel.

CHAPTER III

CHANGES IN MICROHARDNESS OF ENAMEL FOLLOWING TREATMENT WITH LACTATE BUFFER

Earlier studies on the aetiology of dental caries using the artificial mouth by Pigman and co-workers (1954, 1955) were essentially of a qualitative nature. More recently a semi-quantitative method was reported (Caldwell et al., 1958) in which changes in microhardness were used for measuring the rate of carious attack *in vitro*. However, because of the variability of the rate of surface softening, the results from many teeth were averaged in order to establish trends.

This study was undertaken to determine the rate of change of enamel hardness under simpler conditions, with the purpose of demonstrating particularly whether variation in the resistance of teeth was an important factor. The rate of softening of teeth produced by a lactate buffer at a constant temperature and constant pH provides a simpler system than the artificial mouth to test the possible variability of individual teeth. This chapter presents data on the rate of softening of human teeth exposed to 0.001 M lactate buffer at pH 5.0 and 35°C for periods up to 10 hours. The effect of topical fluoride on the rate of softening is also described.

MATERIALS AND METHODS

Human anterior teeth for this study were collected from dentists in Birmingham, Alabama, where the fluoride content of the drinking water is 0.0 to 0.3 ppm. After extraction, the teeth were stored in distilled water and thymol to prevent drying and to inhibit bacterial growth. Prior to measuring the hardness, the enamel surface was cleaned with water and a rubber prophylaxis cup, using very light pressurs.

Hardness determinations were made as described by Caldwell et al. (1957) using a Kentron microhardness tester and a 500 g. load applied for 10 sec. Ten impressions were placed on each enamel surface at the beginning of the experiment and after each treatment period. Because of the non-planarity of intact enamel surfaces, the hardness measurements were made by adjusting the tooth and indenter at each reading and not solely at the first reading as recommended for plane surfaces. This results in an initial impression before the regular impression is superimposed upon it. With the standard metal hardness block, this procedure results in a reduction of the hardness values of about 70 KHN for hardnesses around 600 KHN. For a ground tooth surface, the procedure used in the present work gave an

average hardness of 269 KHN, whereas the recommended procedure gave 318 KHN. The hardnesses for teeth reported previously by Caldwell et al. are then presumably reproducible but somewhat lower than they might have been if the method used for metal surfaces had been possible. The average for these ten impressions was taken as the hardness value. Groups of 4 to 6 teeth at a time were immersed in 1 litre of 0.001 M sodium lactate buffer at pH 5.0 maintained at 35°C and stirred continuously. The acrylic boxes carrying the teeth were sealed to the bottom of the beaker by the use of a sticky wax, so that the experimental surface lay face up. At predetermined intervals, the teeth were removed from the buffer solution, washed in distilled water, and stored in an atmosphere saturated with water until their hardness had been determined. They were then exposed to a fresh buffer solution for an additional period, and the change of hardness determined.

During each experiment, the pH of the buffer was checked frequently; if a deviation of more than 0.2 pH unit occurred, the buffer was replaced by a new solution. This was only found necessary in treatment over the longer time-intervals. These conditions were established after an extensive study of the effect of variations in pH and especially lactate concentrations, and seem to be optimal

for the purpose. At higher concentrations of lactate, the rate of softening proceeded too rapidly, surface loss was apparent, and satisfactory measurements could not be made. Lower concentrations had insufficient buffer capacity to maintain the initial pH.

Ground surfaces were prepared by a modification of the technique described by Hammarlund-Easler (1955) using increasingly fine grits of silicon carbide paper (Minnesota Mining and Mfg. Co., St. Paul, Minn., U.S.A.). Final polishing was effected with emery polishing paper (Norton Berh-Manning, Troy, N.Y., U.S.A.), grit No. 4/0.

In testing the effect of fluoride, the teeth were placed in acrylic boxes immersed for 2 hours in 500 ml. of a 2% solution of NaF at pH 6.5 and 35°C. The fluoride-treated teeth were then immersed in a lactate buffer under the same conditions as for the control group of teeth.

RESULTS

Intact surfaces.

Forty intact teeth were exposed to the lactate buffer during 53 experimental runs and the results are shown in Fig 10. Time intervals of 2,3,4,6,8 and 10 hours were used. Because of the curvature of the tooth surfaces, the number of symmetrical indentations that could be made was limited and usually only two time-periods could be employed for each tooth.

It will be noted that the over-all pattern is a fairly constant linear rate of decreasing hardness of intact enamel over the 10-hour period. In Table VII, the average percentage decrease of Knoop Hardness Number per hour at each time-interval is tabulated.

Similar studies were made of the rate of softening of the intact surface enamel of cattle incisor teeth. The results reported in Table VII show that the softening rate is somewhat greater than that found for human teeth. The cattle teeth are similar to those of human beings but are much larger. Their average initial hardness of 356 $\pm$ 28 KHN for 9 teeth was nearly the same as for human teeth (367 $\pm$ 35 KHN) as measured in this investigation.

Ground surfaces

Twenty-two teeth which had been used for measurements

of the resistance of their intact surfaces were ground and exposed to lactate buffer during 54 experimental runs. For the ground surfaces, the range of values (Fig. 11) was less than with the intact surfaces, and the standard deviation of the individual readings was less. The fall in hardness was again found to proceed at a constant linear rate (Table VII) which was more rapid than that for the intact surfaces.

Effect of fluoride on the softening of enamel surfaces.

The treatment of enamel surfaces with a 2% solution of sodium fluoride produced no significant changes in the absolute hardness. A group of 6 teeth with intact enamel surfaces was found to have an average hardness of 397 KHN and, after the application of sodium fluoride, the average hardness was again found to be 397 KHN. Likewise, a group of 6 teeth with ground enamel surfaces had an average initial hardness of 246 KHN and, after fluoride treatment, the average hardness was 244 KHN. Any changes of the individual teeth in hardness, arising solely from fluoride treatment were within the standard deviation of the initial hardness reading, as was found previously by Caldwell et al. (1958). These hardness changes were both greater and less than the original values and did not indicate a definite trend.

Phillips and Swartz (1948) found a slight increase

in the hardness of ground enamel after fluoride treatment (5.1% with NaF and 7.1 with SnF_2). An <u>in vivo</u> study (Hord and Ellis, 1949) using young dogs, indicated an increase in enamel hardness of 13%, following 8 topical applications of sodium fluoride. The results of the present work do not agree with these earlier reports.

After exposure for 8 hours to a lactate buffer, 14 teeth with intact enamel surfaces showed a decrease in hardness of 5.4% per hour as compared to 5.1% per hour for a group of 9 teeth which had been fluoride-treated (Table VIII). This difference was not significant ($P > 0.1$).

For the ground teeth, the untreated surfaces showed an average decrease in hardness of 7.5% per hour for a group of 10 teeth. On the other hand, the fluoride-treated ground surfaces of 12 teeth softened at a relatively slower rate of 5.8% per hour. This difference was significant at $P < 0.01$.

<u>Hardness of naturally carious areas.</u>

The hardness of a number of naturally carious lesions was also measured. Although suitable areas were difficult to find, some measurements were made, usually on interproximal surfaces. Typical results are shown in Fig. 12 in which hardness measurements across a carious surface are given. Similar hardness measurements were also obtained

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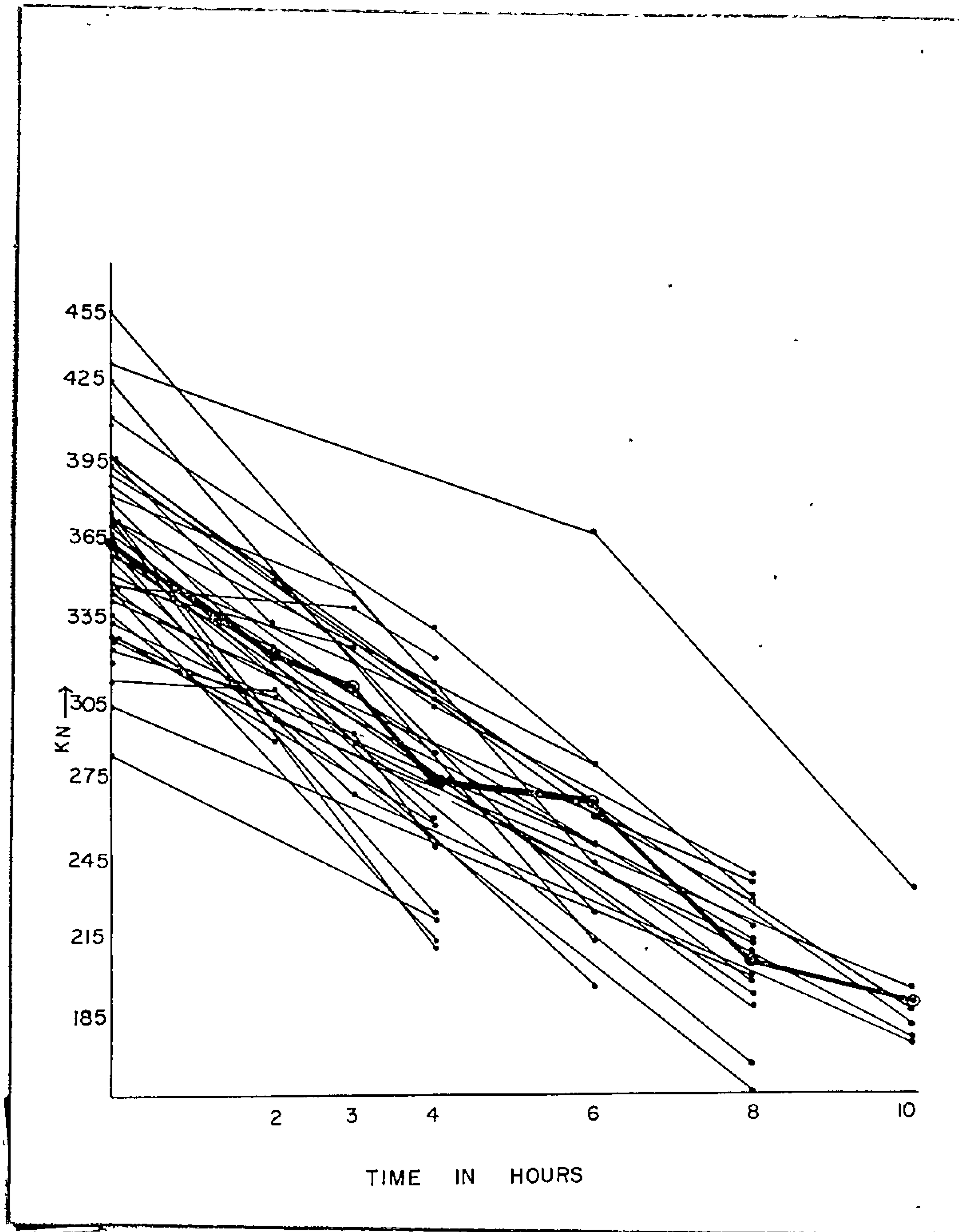
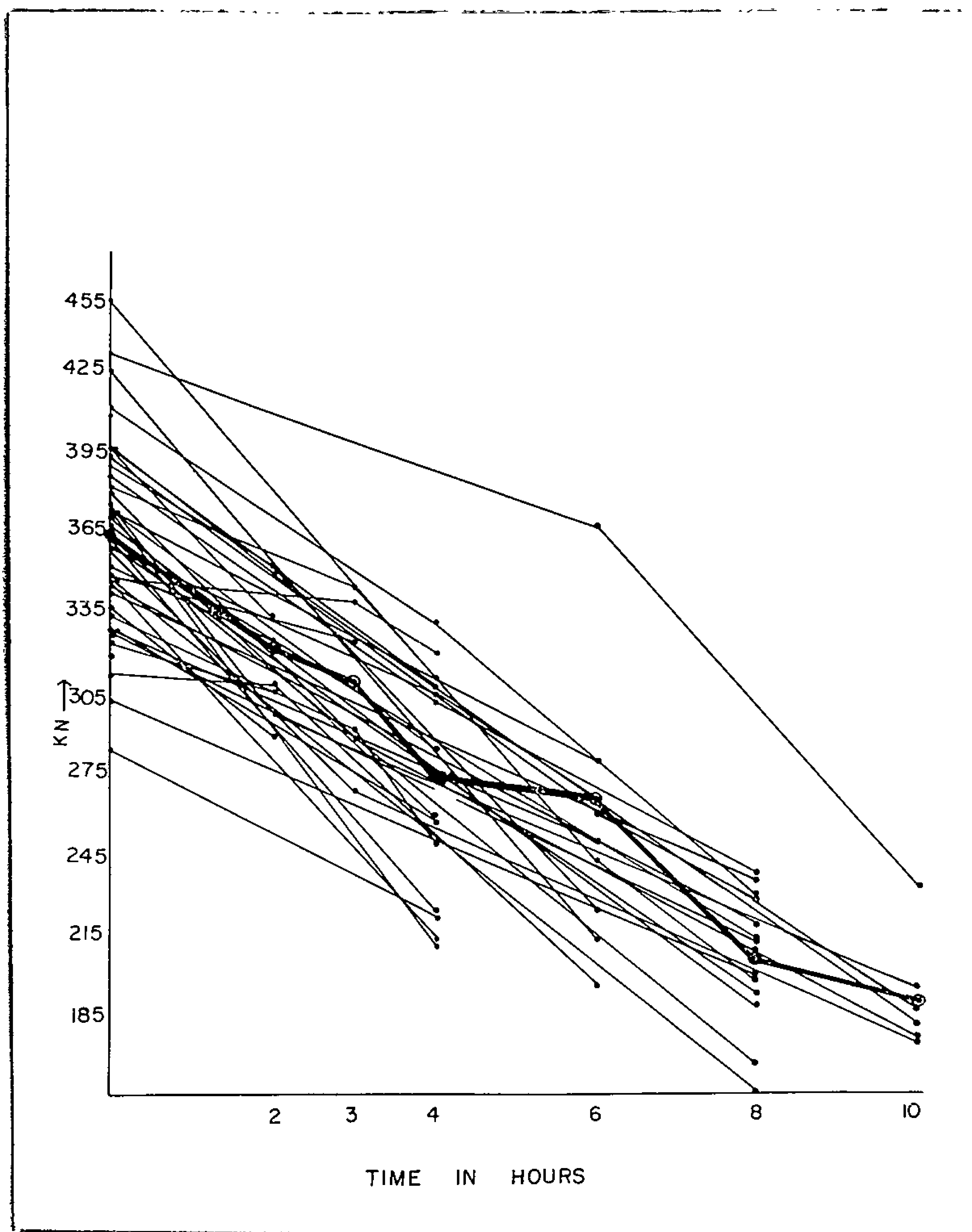


Fig. 10. Decrease in hardness of intact enamel on exposure to lactate buffer. Temperature, 35°C; pH, 5.0; 0.001 M. Light lines indicate individual teeth; heavy line represents average decrease.



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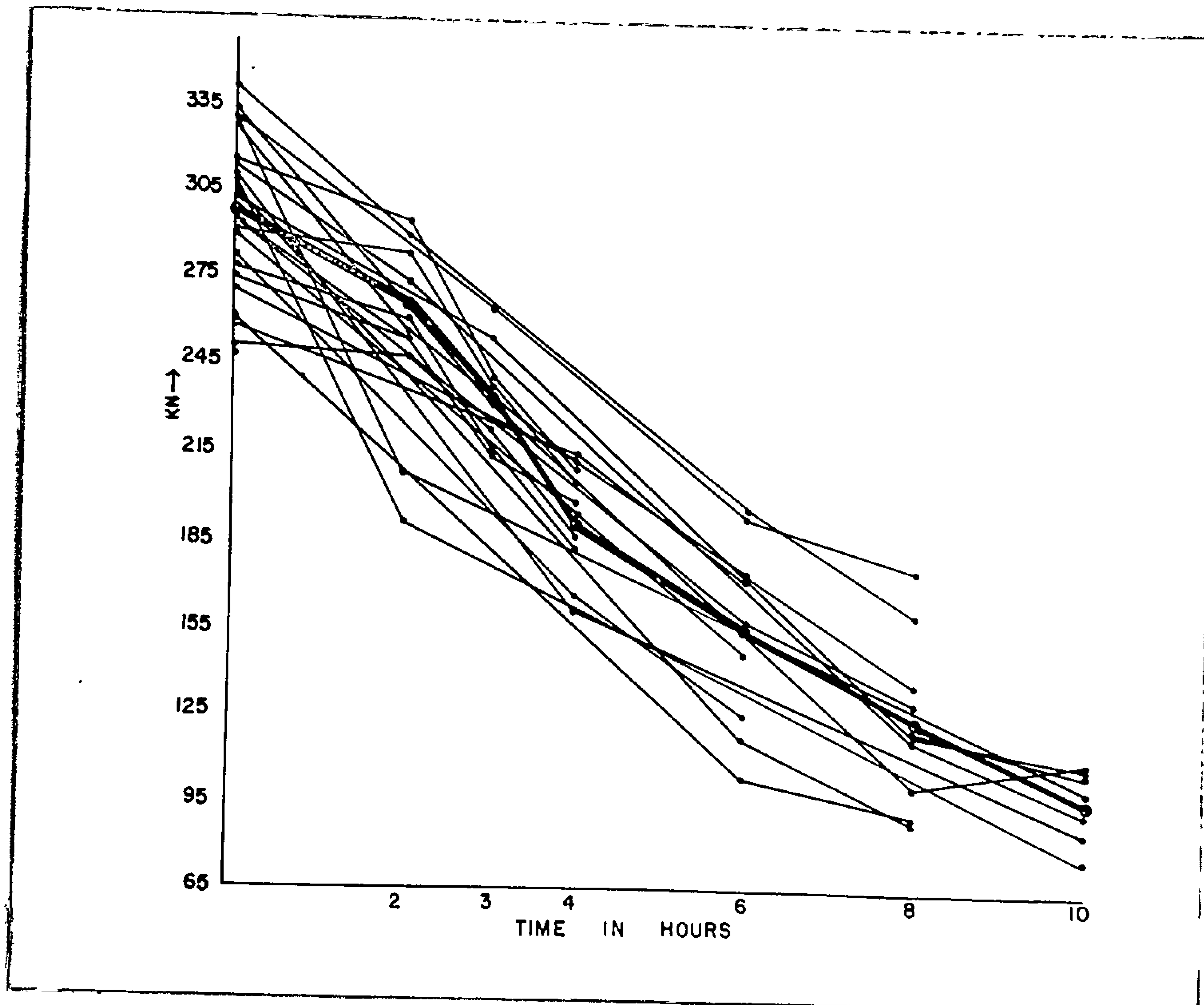

Fig. 11. — Decrease in hardness of ground enamel on exposure to lactate buffer. Temperature, 35°C; pH, 5.0; 0.001 M. Light lines indicate individual teeth; heavy line represents average decrease.

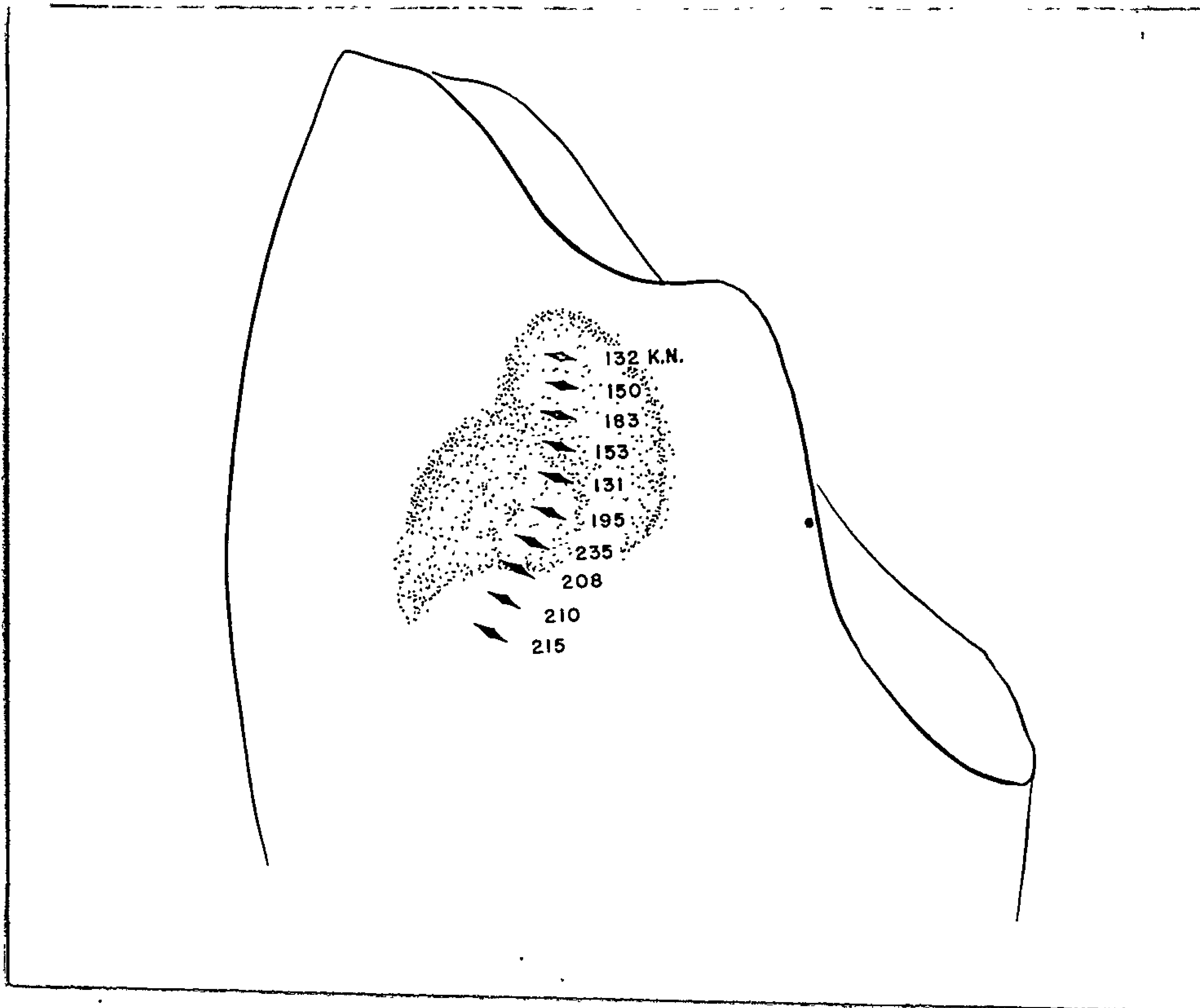

Fig. 12.— Knoop hardness number readings across a natural carious lesion on the interproximal of a canine. The dotted area had a yellowish-white color, but showed no signs of loss of surface.

DISCUSSION

The terms "hard" and "soft" teeth have been used frequently in the dental literature (Miller, 1905). Pickerill (1912) formulated an empirical classification of sclerotic teeth, characterized by hardness and immunity to caries, and malacotic teeth, characterized by comparative softness and susceptibility to caries. In order to determine whether the data of the present work would indicate a correlation between initial hardness and resistance to softening, the initial hardness was plotted against the rate of softening for each tooth at each time-period and the regression lines calculated.

The equations were: $x = 7.9 \times 10^{-3}y + 365$ and $y = 2.9 \times 10^{-5}x + 21$ (x = initial hardness) (y = decrease in KHN per hour), and the corresponding lines were almost perpendicular to each other. As the coefficient of correlation was practically zero ($r = 2.8 \times 10^{-4}$), the initial hardness showed no relation to the rate of decrease of hardness. This finding was similar to that reported by Swartz and Phillips (1952) who found no correlation between the initial hardness of ground surfaces and the amount of phosphorus dissolved by a buffer in a standard time-interval, and to the results of Steel (1955) who found that the surface hardness of enamel and its calcium and

phosphorus solubility at various pH values were unrelated.

The marked tendency for the slopes of the lines for the individual teeth to be parallel (Figs. 10 and 11) also indicates that the rate of softening is independent of the initial hardness. However, although the rates seem to be constant and similar, the harder teeth tend to take longer to reach any arbitrary extent of softening. In this sense, then, hard teeth are more resistant than soft teeth.

The intact tooth surfaces were shown to soften more slowly than ground surfaces. This result is probably related to earlier observations (Brudevold, 1948; Isaac et al., 1958a) that surface enamel is less soluble than the deeper layers. Since the surface layers have been shown to have a high fluoride content and fluoride-treated enamel is less soluble than normal enamel, the lesser rate of softening of the intact surface may arise from the fluoride effect.

The outer enamel layer not only has a much higher concentration of fluoride than the underlying enamel but also appears to accumulate fluoride with age (Brudevold et al., 1956; Isaac et al., 1958b). Teeth used in the present experiments were human anteriors from persons usually older than 30 years and, hence, would be expected to have

accumulated a considerable quantity of fluoride in the surface layers. This accumulation of fluoride may explain the present results (Table VIII) that fluoride treatment had no demonstrable effect on intact surfaces. Previous studies (Myers et al., 1952; Brudevold et al., 1957) using radioactive fluoride indicated that the uptake of fluoride is greatest at ground or etched surfaces or in defective areas. As might be predicted from these studies, the results of the present work showed a significant decrease in the rate of softening of ground enamel which had been treated with fluoride.

The absence of an enhanced resistance of intact enamel surfaces after fluoride treatment as found in the present work may seem inconsistent with the known efficacy of topical fluoride treatments *in vivo*. However, topical treatments are usually made on young teeth, and the teeth in the present work were relatively old. In addition, any enhancement of resistance in adult teeth may be highly localized, since extensive variations of hardness are found in single teeth. These soft "spots" may take up fluoride better than harder areas. Although the resistance of these "spots" to solution may be increased by uptake of fluoride, the increased resistance might not show up in the present measurements which average the

effect over a considerable area. Such soft "spots" are present in unerupted teeth (Caldwell et al., 1958) and presumably represent defective or incomplete development or calcification.

SUMMARY

Human incisor teeth were immersed in 0.001 M lactate buffer at pH 5.0 and 35°C. The rate of softening of the enamel was measured by microhardness tests for exposure times up to 10 hours. A linear rate of softening was found and the softening was expressed as per cent drop of hardness per hour. Cows' teeth gave similar results but the rate of softening was somewhat greater than for human teeth.

Intact enamel surfaces were found to soften at a slower rate than ground and polished surfaces, and the rates were closely similar for all teeth studied. The rate of softening was independent of the initial hardness. Since hard teeth took longer to soften to a given hardness value, hard teeth may be considered to be more resistant than soft teeth.

Prior exposure of enamel to fluoride solution reduced the rate of softening of ground and polished surfaces but not of intact surfaces.

The results suggest that the effects of buffers are similar to true carious attack on enamel.

CHAPTER IV

THE HARDNESS OF FLUOROSED ENAMEL

Most previous studies have been concerned with the hardness of normal or carious enamel, but recently there have been several papers dealing with effect of topical fluoride applications, acids and artificial caries on enamel hardness by Phillips and Swartz (1948), Caldwell et al. (1958) and Newbrun et al. (1959). However, no attempt has been made to evaluate the effect on enamel hardness of fluoride taken systemically during the period of tooth development. Bowes and Murray (1936) noted on grinding fluorosed enamel that it seemed softer than normal. The purpose of this investigation was to measure quantitatively the hardness of fluorosed enamel and determine if any correlation exists between the hardness, the degree of dental fluorosis and the quantity of fluoride in the water supply.

MATERIALS AND METHODS

Thirty permanent anterior teeth were collected from Alamosa, Col., Colorado Springs, Col., Doland, S. Dakota and Post, Texas, where the water supply contains 1.5, 2.6, 2.9 and 5.0 ppm of fluoride respectively. Only teeth from persons who had lived continuously in these locations were included. These teeth were cleaned and mounted in hard inlay wax in individual acrylic resin boxes. The hardness measurements were made using a Kentron microhardness tester with a 500 g. load applied for 10 sec. The procedure outlined for intact surfaces in the previous chapter was observed in determining the Knoop Hardness Number of a tooth.

RESULTS

A total of 344 hardness measurements were carried out on the intact surface of thirty teeth used in this experiment. At least ten readings were taken of each tooth, and the average was taken as the Knoop Hardness of the tooth. These values, together with the degree of mottling, the age group, the fluoride concentration of the water supply and the area are shown in Table IX.

A variable degree of mottling was observed in the teeth used in this experiment. Some teeth appeared normal, others were white and lacking in translucency or showed light or dark brown areas (Figure 14). In assessing the degree of mottling, the classification of Venkateswarlu et al. (1952) was used (see Table X). The Dental Fluorosis Index (D.F.I.) was obtained by totalling m (the grade of mottling), dividing by the number of teeth studied and multiplying by a hundred. The D.F.I. closely resembled the numerical weighted index of clinical severity suggested by Dean (1942), but was on a tooth-unit basis per hundred teeth instead of on a child-unit basis. However, both these fluorosis indices are at best, semiquantitative measurements. Figure 13 compares the D.F.I. with the Knoop Hardness of each of the groups studied. It will be noted that there is an inverse linear relationship.

The Knoop Hardness of the nine teeth from Post, Texas was 305 ± 24 which is significantly lower ($P < 0.01$) than values of 365 ± 35 and 367 ± 35 obtained (Caldwell et al., 1957; Newbrun et al., 1959) for anterior teeth from Birmingham, Ala. where the fluoride content in the drinking water is only 0.00 - 0.03 ppm.

Of the ten teeth from Doland, S. Dakota the average Knoop Hardness was 355 ± 20 which is not significantly ($P > 0.10$) different from the hardness observed for teeth from Birmingham. Unfortunately only three teeth were available from Colorado Springs, Col. and all were from the same patient (55 years old). They were all moderately mottled and quite soft with a Knoop Hardness value of 268 ± 34. While it is considered that this is the correct value of the teeth examined, it would appear that these teeth are not entirely representative of Colorado Springs. Dean (1942) studied 404 children in the 12-14 age group from this area and reported an index of fluorosis of 1.3 which would correspond to about 130 D.F.I. The D.F.I. of the teeth whose hardness was measured was 200, which indicates a greater amount of mottling than is usual for this area. In Colorado Springs the water contains 2.6 ppm of fluoride.

Eight teeth from Alamosa, Colorado (1.5 ppm of fluoride in the drinking water), averaged 396 ± 18 in hardness. This

value is not significantly ($P > 0.10$) different from that of Birmingham teeth (367 ± 35). Only one of this group of teeth showed any evidence of mottling.

Almost all fluorosed teeth exhibited pronounced perikymata, much more than had been noted on the surface of non-fluorosed enamel. These irregularities added to the difficulty of obtaining good hardness indentations.

As a further basis of comparison between high and low fluoride areas, the microhardness of teeth from Binghamton, New York (0.05 - 0.15 ppm F) and Schenectady, New York, (0.05 - 0.1 ppm F) was measured. These are areas where the drinking water is low in fluoride, like Birmingham, and the teeth from these localities showed no evidence of mottling. The results are summarized in Table XI from which it will be seen that enamel hardness in the low fluoride areas differs only very slightly, being 367 ± 35 for Birmingham, Alabama, 369 ± 30 from Schenectady, New York and 374 ± 30 for Binghamton, New York.

TABLE IX.

Hardness and mottling of enamel surface in relation to fluoride in the drinking water

Tooth No.	Knoop Hardness Number ± S.D.	Age	Degree of Mottling	
F1	333 ± 31	20-29	2 m	
F2	291 ± 13		2 m	
F3	323 ± 37		2 m	Post, Texas
F4	322 ± 42		4 m	F.Conc.of water Supply, 5 ppm
F5	322 ± 17		2 m	
F6	314 ± 33		2 m	
F7	334 ± 29	30-49	0 m	
F8	335 ± 23		2 m	
F9	269 ± 43		1 m	
Avg	305 ± 24		DFI* 189	
F10	341 ± 27		0 m	
F11	324 ± 25		2 m	
F12	322 ± 70	30-49	4 m	
F13	350 ± 15		4 m	Doland, S.Dakota
F14	387 ± 41		0 m	
F15	372 ± 27		0 m	F.Conc.of Water Supply, 2.9 ppm
F16	350 ± 25		1 m	
F17	359 ± 36	20-29	0 m	
F18	367 ± 14		0 m	
F19	373 ± 82		0 m	
Avg.	355 ± 20		DFI 110	
F20	220 ± 14		2 m	Colorado Springs, Colorado
F21	298 ± 18	55	2 m	F.Conc.of Water
F22	287 ± 21		2 m	Supply, 2.6 ppm
Avg.	268 ± 34		DFI 200	
F23	400 ± 34		0 m	
F24	383 ± 44	30-49	0 m	Alamosa, Colorado
F25	417 ± 54		0 m	
F26	409 ± 61		0 m	F.Conc. of Water Supply, 1.5 ppm
F27	379 ± 16		0 m	
F28	384 ± 36	20	4 m	
F29	423 ± 39		0 m	
F30	372 ± 32		0 m	
Avg.	396 ± 17		DFI 50	

*Dental fluorosis index (DFI)

TABLE X.

Grade of mottling	Description of mottling	Valuation
Normal	No mottling	0 m *
Mild	White opacities or patches on the enamel; very faint line (yellow) across the enamel	1 m
Moderate	Distinct brown stain becomes well established	2 m
Severe	Beside the well established brown line, the tooth is worn out, edges are chipped off and there is considerable pitting all over the enamel	4 m

*m is the unit of manifestation of fluorine in the form of mottled enamel.

TABLE XI.

Average hardness of enamel surface in relation to drinking water, comparing areas with low and high fluoride concentration.

No. of teeth tested	Average Knoop Hardness ± S.D.	F.conc. of water supply in ppm	Area
40	367 ± 35	0.00–0.03	Birmingham, Alabama
8	374 ± 30	0.05–0.15	Binghamton, New York
10	369 ± 30	0.05–0.1*	Schenectady, New York
8	396 ± 18	1.5	Alamosa, Colorado
3**	268 ± 34	2.6	Col.Springs, Colorado
10	355 ± 20	2.9	Doland, S.Dakota
9	305 ± 24	5.0	Post, Texas

*Schenectady started fluoridation of its water supply in 1952 but due to technical difficulties it has only been carried on intermittently and for a majority of the time it was untreated. As the teeth in this group were all from individuals over 30 years in age, the effect, if any, would have been due to topical rather than systemic fluoride.

**All three teeth in this group were obtained from the same individual (55 years old) and showed pronounced mottling.

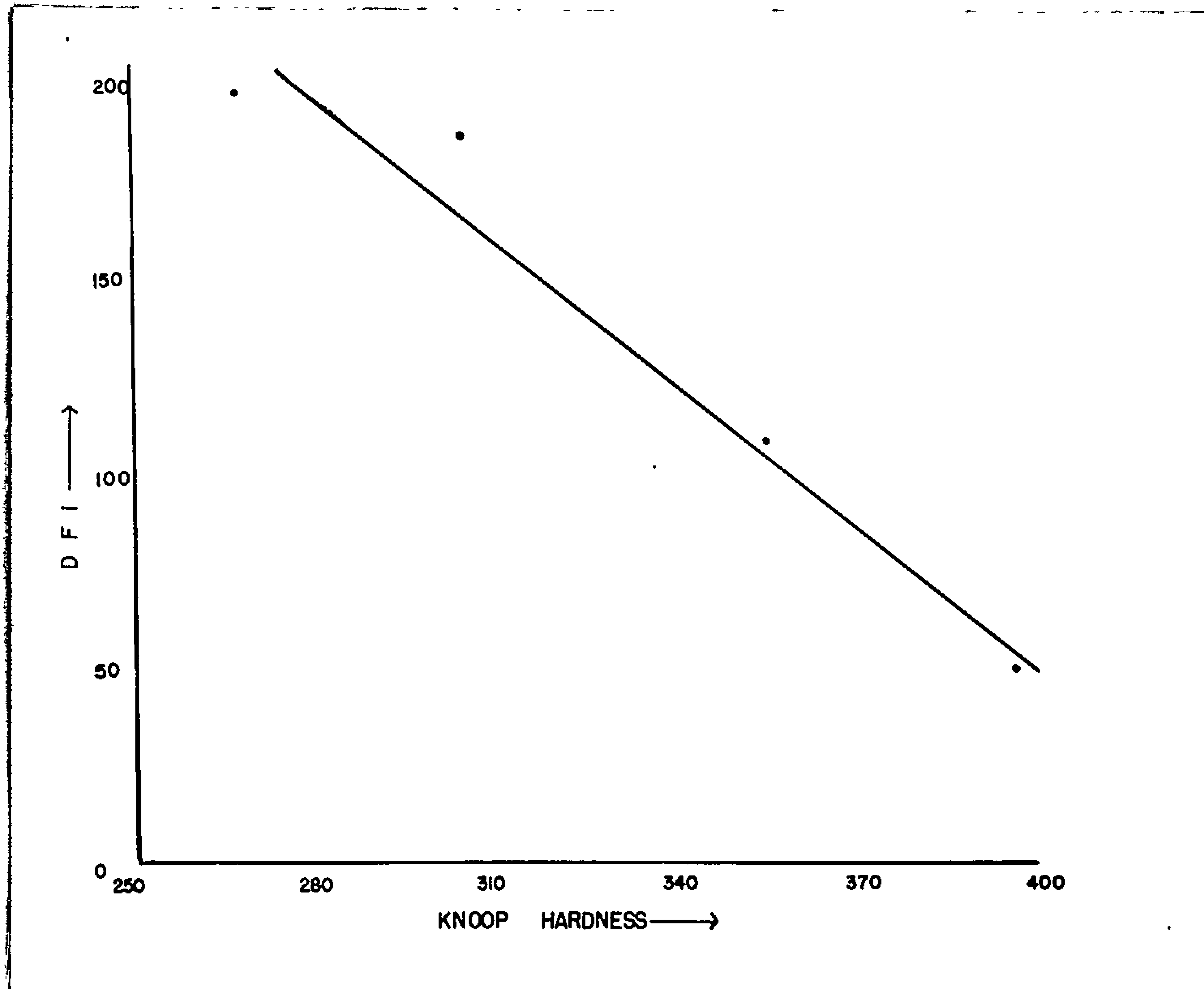

Figure 13

Relationship of mottling (Dental Fluorosis Index) to Knoop Hardness number of the enamel.

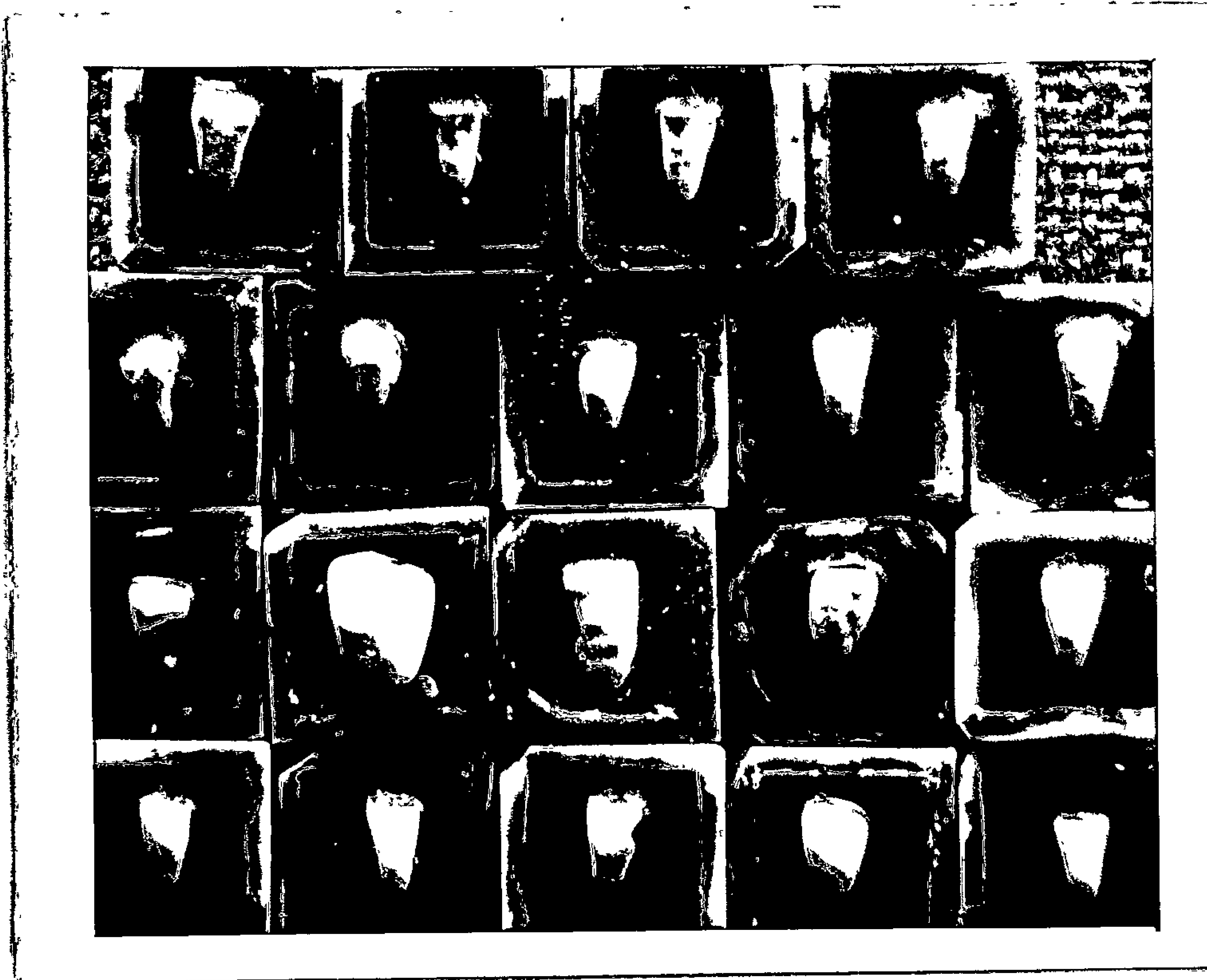

Figure 14. Photograph of teeth F1 - F19 (Table IX) illustrating the degree of mottling.

DISCUSSION.

The classical epidemiological findings of Dean (1942) have shown an extraordinarily precise quantitative relationship between the degree of dental fluorosis and the fluoride concentrations of the public water supply. Subsequent investigations revealed that an inverse relationship also existed between the fluoride content of the water and the dental caries experience of children who consumed the waters throughout tooth development. As an extension of these epidemiological findings, it was shown (Isaac et al., 1958) that the amount of fluoride deposited in the enamel during tooth formation and posteruptively was directly proportional to the fluoride content of the water supply and further than enamel solubility was inversely proportional to its fluoride content. The present investigation has shown another correlation, namely the greater the degree of dental fluorosis the lower the Knoop Hardness of the enamel.

These findings appear at first to be inconsistent with established data showing an inverse relationship between fluoride content and enamel solubility. It is a popular misconception that solubility and hardness run parallel. However, the studies of Swartz and Phillips (1952) showed no correlation between the initial hardness of enamel and the amount of phosphorus dissolved by a buffer in a standard time

interval. Also our previous studies (Newbrun et al., 1959) found no relation between the initial hardness of enamel surfaces and the rate of decrease of hardness. There have been attempts at various times, Richter (1931) and Hodge (1939), to establish a relationship between hardness and caries susceptibility. However, it was learned that the microhardness of sound enamel and dentine does not differ on the average from the microhardness of sound portions of enamel and dentine from carious teeth and that the hardness was independent of the amount of caries in the mouth. It is apparent then, that fluorosed teeth can be softer than normal teeth and still remain more resistant to decay.

It has been reported in the literature that a topical application of a fluoride solution to an enamel surface will result in an increased hardness. Phillips and Swartz (1948) found aan increase of 5.1% with sodium fluoride and 7.1% with stannous fluoride in the hardness of ground enamel following *in vitro* treatment. Similarly Hord and Ellis (1949) claimed an increase in enamel hardness of 13% following topical applications of sodium fluoride to the teeth of dogs *in vivo*. Recently Herrmann (1958) and Herrmann and Rozeik (1959) have described a twofold effect (topical and systemic) on the teeth of albino rats following the feeding with very high concentrations of fluoride, 500 ppm or 1000 ppm, in the

diet. They noted an increase in hardness of molars already formed and a decrease in hardness for incisor teeth which were still forming. The higher the fluoride concentration the lower the hardness of developing teeth. Herrmann suggested that in systemic fluoride ingestion, calcium fluoride was deposited during mineralization, and this was softer than hydroxy-apatite. He explained the increase in hardness of enamel which had already calcified as arising from the formation of fluorapatite. On the other hand, Bibby and Brudevold (1954) in reviewing the mechanism of fluoride fixation following topical treatment pointed out that high fluoride concentrations result in deposition of fluoride as calcium fluoride. Lower concentrations of fluoride as occur in the environment of the teeth during formation would lead to deposition of fluorapatite, not calcium fluoride, even in severely mottled enamel. Careful studies described in the previous chapter have failed to reveal any change in hardness after topical fluoride treatment (Newbrun et al., 1959). These results, while not clarifying the situation concerning the effect of topical applications of fluoride on enamel hardness, indicate that high amounts of fluoride taken systemically during the period of tooth development do affect the hardness of enamel.

It is interesting to note that this systemic effect of

fluoride on enamel hardness does not become apparent until levels in the water supply exceed 1.5 ppm F. The teeth from Alamosa, Colorado (1.5 ppm F in the drinking water) were harder, though not significantly, than the teeth from the low fluoride areas and significantly harder than teeth from very high fluoride areas. This suggests that there may be an optimal level of fluoride concentration for enamel hardness as there is for dental caries and mottling, and that this value lies somewhere between 0.15 ppm and 2.6 ppm F.

SUMMARY

The microhardness was measured on 30 teeth collected from Alamosa, Colorado; Colorado Springs, Colorado; Doland, S. Dakota, and Post, Texas where the water supply contained 1.5, 2.6, 2.9 and 5.0 ppm of fluoride, respectively. The average hardness of teeth from each of these areas was compared with the average hardness of teeth from Birmingham, Alabama (0.00 - 0.03 ppm F in the drinking water). It was found that teeth from Colorado Springs and from Post were significantly softer than those from Birmingham but that the teeth from the other locations did not differ significantly. The degree of dental fluorosis appeared to be inversely proportional to the Knoop Hardness.

CHAPTER V.

GENERAL CONCLUSIONS.

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It is doubtful whether any single technique, be it chemical, physical, histological or biological, will provide the ultimate explanation of the calcification and decalcification mechanisms. The pooling of data derived by a variety of methods has permitted rapid progress in recent years, and it is hoped that these studies will further this progress. Microhardness testing is a suitable means of examining the resistance to plastic deformation of sound, carious, fluorosed and artificially softened enamel. Indirectly the hardness appears to be a measure of the degree of mineralization.

One of the difficulties in assessing previous hardness observations has been a lack of means of cross comparison of data derived by different laboratories. Considerable confusion would be avoided if the multiplicity of types of hardness measurements could be reduced and one particular scale adopted for general use. The information assembled in Chapter I should aid in relating the results of various hardness investigations. It must be recognised however, that an absolute comparison of relative hardness can only be obtained from tests made by one observed on the same apparatus used under identical conditions of loading and measurement of impression size.

Nevertheless, it was noted, in Chapter II, that one observer using two different types of instruments but with similar shaped indenter and identical load could obtain closely related values. The different hardness numbers for enamel quoted in the literature could therefore not be attributed solely to differences in instruments.

A fall in hardness between intact surface enamel and ground and polished subsurface enamel has been reported and the data in this thesis corroborate these findings. It was also found that hardness was significantly affected by testing sections in different planes and different locations in the enamel of the same tooth. These changes in hardness could be attributed to alterations in the architectural arrangement of the enamel prisms and their matrix in different planes. It was concluded that many previous tests reporting a low hardness for enamel appear to have been made in a plane where fracturing and cleavage along the prisms is greatest.

Intact surface enamel is considerably harder than most currently used dental restorative materials.

In view of earlier solubility studies, it was not surprising to learn that intact enamel softens more slowly than subsurface enamel. An interesting discovery, however, was that under uniform conditions the rate of softening was

independent of the initial hardness and was closely similar for all teeth studied.

Topical fluoride treatment did not alter the hardness of enamel. However, it significantly reduced the rate of softening of ground and polished surfaces although not of intact surfaces. Fluoride taken systemically in excessive amounts (5 ppm F) produced teeth whose enamel was significantly softer than the enamel of control teeth from Birmingham, Ala. (0.00 - 0.03 ppm F). Systemic fluoride taken in optimal or near optimal amounts did not result in any significant difference in hardness from the control teeth of a low fluoride area. The degree of dental fluorosis appeared to be inversely proportional to the Knoop Hardness.

These data are not interpreted as vitiating the well established epidemiological findings as to the efficacy of topical fluoride applications or systemic fluoride intake in optimal amounts in reducing the dental caries experience. However, the mechanism involved is not one of "hardening" the enamel as has been suggested but rather of reducing the rate at which enamel softens.

CHAPTER VI.

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