PHOSPHATE EXCHANGE IN HYDROXYLAPATITE, ENAMEL, DENTIN, AND BONE

II. EFFECT OF FLUORIDE ON THE EXCHANGE

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In previous studies of the reactions of fluoride ion with synthetic hydroxyapatite and powdered enamel and dentin (1, 2), it was thought that low concentrations of fluoride ion might be affecting the rate of recrystallization of the apatites. Neuman et al. (3) found that fluoride impregnation had no effect on calcium and phosphate exchange of glycol-ashed bone.

The experiments described below were designed to study the effect of various amounts of fluoride on the phosphate exchange of various mineralized tissues and synthetic hydroxylapatite. The fluoride was introduced in the buffer solutions, and some of the tissues studied were previously fluoridated either in vitro or in vivo.

EXPERIMENTAL

Details of the phosphate exchange experiments with 0.005 M KH$_2$PO$_4$ were given in Paper I of this series (4). Samples of synthetic hydroxylapatite, human enamel and dentin, and rat dentin and femur, including samples fluoridated in vivo, were obtained from studies reported previously (1, 2, 5). Tissues treated in vitro were exposed to an aqueous solution of NaF containing the indicated fluoride concentration for a period of 1 month at 37°C.

Results

Fig. 1 shows the effect on P$^{32}$ exchange of treating rat incisor dentin with 2, 10, and 100 p.p.m. of fluoride in the buffer as compared with use of buffer containing no fluoride. The curves obtained from 2 and 10 p.p.m. of F are higher than the control curve where no fluoride was used, indicating a more rapid rate of P$^{32}$ exchange for the first 30 days and resulting in a larger over-all amount of exchange. However, with 100 p.p.m. of fluoride less exchange was obtained than when no fluoride was used.

Similar results were obtained when hydroxylapatite, enamel, human dentin, and rat femur were employed, although the over-all amount of $^{32}P$ exchange varied considerably, depending on the tissue. The final exchange after 4 months with 2 and 10 p.p.m. of F was always higher than with 100 p.p.m. of F, although their relative positions were sometimes reversed. With 100 p.p.m. of F the exchange was sometimes greater than when no

![Diagram](http://www.jbc.org/)
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fluoride was used, but the final exchange was always less than when either 2 or 10 p.p.m. of F were present in the buffer.

Fig. 2 shows the effect of fluoride already present in the tissues added either in vivo or in vitro. The tissue treated in vivo contained 0.52 per cent fluoride, the tissue treated in vitro, 0.87 per cent fluoride. There is an initial increase in the rate of exchange and then a decreased amount of exchange as the reaction proceeds. The results were similar with human dentin treated in vitro with 2 and 100 p.p.m. of fluoride. Human enamel treated in vitro and rat femur treated both in vivo and in vitro exchanged less than the controls throughout the entire exchange period.

Fig. 3 shows that fluoride in the buffer increases the rate and amount of exchange of tissues previously treated with fluoride in vitro. A similar effect was obtained with tissues previously treated with fluoride in vivo.

DISCUSSION

When fluoride is added to the buffer, it has a slight effect during the initial reaction which is restricted almost entirely to surface exchange. In most cases the effect is a slight increase in the amount of exchange. As the reaction proceeds, the exchange with hydroxylapatite and enamel must be through recrystallization. In the case of dentin and bone there is probably a slow surface exchange as the solution diffuses through the organic matrix (4). This is combined with some recrystallization, and still later the exchange is due entirely to this latter effect. In all cases as recrystallization begins to take place, the fluoride increases the rate and amount of exchange to a considerable extent. It seems probable that a fluorapatite crystallization center, in contrast to the hydroxylapatite, would induce a greater stability to the crystal, which would tend to increase the recrystallization of the particles because of the tendency to reach the most stable thermodynamic state.

The increase in the rate of exchange of P32 caused by increasing fluoride concentrations falls off markedly at 100 p.p.m. of F. At higher sodium fluoride concentrations the sodium ion may substitute for calcium ions (6), possibly accounting for the decrease at a concentration of 100 p.p.m. of F. Thus sometimes the 2 p.p.m. of F and sometimes the 10 p.p.m. of F curve are uppermost, showing the greatest amount of exchange, but the 100 p.p.m. of F curve is always lower and sometimes even drops below the 0 p.p.m. of F control curve.

With fluoride already present in the tissues, the exchange rate sometimes increased slightly at first, but the rate of subsequent exchange was less than that of the control, probably because of decreased solubility. Recrystallization because of the fluoride has been completed before addition of the P32 and the effect of the fluoride ions is to decrease the solubility, slowing,
rather than increasing, the $P^{32}$ exchange rate. This increased exchange during the first 24 hours may be caused by a release of fluoride ions into the solution. This would correspond to the increased exchange of $P^{32}$ with previously fluoridated tissues when additional fluoride is added to the buffer.

In a previous paper (5) the similarity in behavior of dentin and femur treated in vivo and in vitro with fluoride in respect to solubility and hydroxyl exchange was given as evidence for the formation of fluorapatite in vivo. The similarity in $P^{32}$ exchange of the tissues fluoridated in vivo and in vitro is additional evidence for the formation of fluorapatite in mineralized tissue when fluoride ion is ingested.

SUMMARY

The simultaneous use of fluoride ion and $P^{32}$ has shown that fluoride ion increases the rate of exchange of $P^{32}$ with synthetic hydroxyapatite, human enamel and dentin, and rat dentin and bone. The rate is also increased by the presence of fluoride ion in the buffer when enamel, dentin, and bone have been previously treated with fluoride either in vivo or in vitro. The rate of exchange of $P^{32}$ with previously fluoridated enamel, dentin, or bone, compared with the corresponding untreated control, is decreased.

Additional evidence for the formation of fluorapatite in mineralized tissues has been presented.

BIBLIOGRAPHY

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