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SMALL QUANTITY FIELD DISINFECTION

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Globaline and halazone have been used as disinfectants since the second World War. Currently, both are undergoing re-evaluation to find a disinfectant tablet that can dissolve fast and still pass the tests of storage and time.

THERE are many occasions when water disinfection must be practiced on a small scale and under adverse conditions. The need for a ready-to-use disinfectant is greatest during military operations or at times of natural disasters when small groups of people or even individuals have to depend upon sources of water which might be contaminated. Even during peace time, campers, sportsmen and adventurers have need for a packaged, instant-disinfectant such as a tablet. This need was first recognized more than 50 years ago.[1] However, only a few preparations, usually containing either iodine or chlorine, were in use at the beginning of the Second World War. At that time a team of scientists and engineers conducted an extensive investigation at Harvard University under a contract with the Committee on Medical Research of the Office of Scientific Research and Development. The Harvard researchers [2] listed the following desirable properties of chemical disinfecting agents which are intended for use under field conditions:

1. The disinfectant should be made available as a tablet of such size as to permit use of a single or at most two tablets for a small quantity of water.
2. The method of application should be simple, substantially foolproof, and not unduly time consuming.
3. The tablet should disintegrate or dissolve quickly and liberate its active ingredient rapidly in order to allow as much time as possible for the kill.
4. Disinfectant dosages should be sufficient to ensure disinfection of all kinds of natural waters without testing for residual concentrations of the disinfectant.
5. The treated water should be acceptable to the user. Its odor, taste and appearance should not be objectionable and foods and beverage powders or concentrates placed in the water should not be changed in normal appearance or flavor.
6. The treated water should not be toxic or otherwise undesirably physiologically active over reasonable periods of use. The water, furthermore, must not interfere with essential prophylactic or therapeutic medication.
7. The treated water should not be corrosive to water containers.
8. The disinfecting agent should be stable under conditions of storage and actual use.
9. The ingredients required in compounding the disinfectant should be economically and strategically available.
10. Manufacture of the chemical agent should lend itself to large scale preparation with normally available chemical and pharmaceutical equipment.

Ingredients of a Disinfecting Tablet

In addition to a disinfectant, a tablet must contain substances which aid in its manufacture, its dissolution, and which confer buffering properties which promote the process of disinfection. These ingredients may be classified as follows:

Filler. A filler or an excipient is always required in pharmaceutical practice to give the tablet adequate bulk. A number of fillers are available but a disinfecting tablet must employ one which is soluble in order to preserve the clarity of the treated water. At the same time, the filler should not be hygroscopic and should be inert to the disinfecting chemical. As shall be discussed later, the halazone tablet has sodium chloride as an excipient. A number of soluble nitrates, phosphates, acetates and sulfates may also be employed.

Buffer. The selection of a buffer to promote disinfection is very important. For any agent which releases chlorine, it is desirable that the pH of the chlorinated water be less than 8. Above pH 8, the predominance of hypochlorite ion seriously reduces the disinfection capability. Similarly, when iodine is used, the pH of the solution should not be less than 7. Otherwise, the viricidal efficiency of hypoiodous acid will be sacrificed. Often, the buffer will also serve as a filler. This is true of globaline tablets which employ disodium dihydrogen pyrophosphate as a buffer as well as an excipient.

Lubricant. The function of this ingredient is to lubricate the punches of tablet-making machines. Talc is a popular lubricant. Calcium or magnesium stearate are also sometimes added. The function of a lubricant may sometimes be performed by the filler.

Swelling Agent. Certain colloidal clays, such as bentonite, promote the disintegration of tablets by swelling in water and causing the tablet to burst. The clay is chemically inert but physically very active.

Test Organism

The disinfecting agent employed for waters obtained under emergency conditions must be capable of killing the most resistant waterborne pathogen. The Harvard Report [2] states that "Leaving out of consideration the virus of infectious hepatitis, the cysts of *Endamoeba histolytica* appear to be the most resistant waterborne pathogens that must be dealt with in the water disinfection and so appear to determine the pattern of accomplishment that must be established both in the laboratory and in the field." Much work has since been carried out on various enteroviruses and the results confirm the earlier observations that cysts of *Endamoeba histolytica* offer greater resistance than any enteric virus, including infectious hepatitis, to the disinfecting action of chlorine.[2] Morris,[3] for instance, quotes other investigators who state that the concentrations of HOCl needed to achieve a 99 per cent kill in 10 min at 5°C for virus and cysts are 0.002–0.4 ppm and 10 ppm respectively. Chang [4] presents

data for iodine which shows that, for a contact period of 10 min at 18°C and for a 99.9 per cent kill, the concentrations of I_2 and HOI needed for poliovirus Type I and *E. histolytica* are those given in Table 1. Table 1 indicates that, for effective disinfection of cysts and virus with iodine, both molecular iodine and hypoiodous acid should be present in solution. At pH 7, a dilute solution of iodine contains almost equal percentages of molecular iodine and hypoiodous acid.[5]

Tablets in Use

There are currently two tablets being used for water disinfection in the United States. The halazone tablet has been in use prior to and during World War II. The disinfectant employed is a chlorine compound. The other tablet, globaline, which contains an iodine-based disinfectant, is used by the US Armed Forces for the disinfection of canteen waters.

The composition of the halazone [2] tablet is given in Table 2.

The chemical name of halazone is p-dichlorosulfonamidobenzoic acid. It reacts with water to release hypochlorous acid up to 50 per cent of the titrable chlorine present. One tablet dissolved in a quart of water liberates a titrable chlorine concentration of 2.3 ppm and a maximum concentration of HOCl (as Cl_2) of 1.1 ppm. Titrable chlorine is defined [2] as the total oxidizing power of the material or solution under consideration which is effective in oxidizing iodine ion to iodine in dilute acetic acid solution, expressed as ppm of elemental chlorine. The reaction of the halazone in water is:

$$Cl_2NO_2SC_6H_4COOH + H_2O \rightarrow ClHNO_2SC_6H_4COOH + HOCl$$

In this tablet sodium chloride is the filler and the remaining two compounds form an alkaline buffer.

The globaline tablet derived its name from a chemical compound which was developed at Harvard. Globaline was first identified as triglycine hydroperiodide, $(NH_2CH_2COOH)_3:HI:I_2$ (2). The formulation was later modified to tetraglycine hydroperiodide $(NH_2CH_2COOH)_4:HI:1.24I_2$. This compound provides 42.32 per cent titrable iodine and 59.42 per cent total iodine. The composition of the globaline tablet is given in Table 3. One tablet dissolves in a quart of water to give 8 ppm of titrable iodine.

The talc is employed as a lubricant and disodium dihydrogen pyrophosphate serves as an acid buffer as well as an excipient. This acid buffer serves to lower the pH of natural waters for, at the time the tablet was developed, it was believed that elemental iodine was more germicidal than its main hydrolysis product, hypoiodous acid.

TABLE 1

Concentrations of I_2 and HOI Needed for Poliovirus Type I and E. Histolytica

Species	Poliovirus Type I	*E. histolytica*
Iodine, mg/l	20	2.5
Hypoiodous acid, mg/l	0.45	4

TABLE 2

Composition of Halazone Tablet

Component	Amount *mg*
Halazone	5.30
Soda ash, dried	5.18
Boric acid	11.92
Sodium chloride	114.00
Weight/tablet	136.40

Comparison of the Globaline and Halazone Tablets

An exhaustive study of the properties of both globaline and halazone has been conducted.[2] The following is a summary of some of the results:

Dissolution Time. Field studies employing soldiers in acceptability tests indicated that they considered rapid solubility of tablet as a primary criterion for acceptability. They were impatient with agents that required a waiting period of more than 10 min.

For field simulation, the tablet to be tested was placed in a liter volumetric flask containing tap water at 23°C. The stoppered flask was then inverted end-over-end continuously, causing the tablet to drop through water until it was dissolved. These tests showed that while globaline disintegrated and dissolved in less than one min, standard halazone tablets dissolved in 7.5 min. Thus, in the case of halazone, the actual contact time between the disinfectant and the organism would be 2.5 min if 10 min is taken as the total waiting time.

The disintegration of the tablets is primarily controlled by the filler and expanding agent used in the tablet. Since halazone contains sodium chloride which hardens or "sets up," it suffers from a low rate of solution. The solubility of halazone itself varies with pH. It is low and constant up to a pH of about 4. Above this pH, the solubility increases rapidly, either because of hydrolysis of the dichlor group or through ionization of the carboxylic acid group.

pH	3.8	5.5	5.6
halazone solubility—g/l	0.09	0.83	1.200

It is possible that a change in the filler now employed in halazone might improve the dissolution time of the tablet markedly.

TABLE 3

Composition of Globaline Tablet

Component	Amount *mg*
Tetraglycine hydroperiodide	19.3 to 21
Disodium dihydrogen pyrophosphate ($Na_2H_2P_2O_7$)	82.5 to 92.3
Talc	not more than 6
Weight/tablet	110 to 120

The solubility of globaline is far greater than that of halazone; about 380 g/l of distilled water. The disintegration of the tablet, however, limits the rate of solution of the globaline.

Decreased water temperature increases the time required for solution for both tablets in accordance with the Van't Hoff-Arrhenius formulation. Some of the dissolution times observed were:

	10°C	20°C	30°C
Globaline	1.9 min	1.2 min	0.8 min
Halazone	9.5 min	8 min	6.5 min

In general, tests showed that storage at 60°C and room humidity did not affect the solution properties of either tablet.[2]

Comparing iodine and chlorine as disinfectants for small water supplies,

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the former appears to have certain advantages for the following reasons:

1. On a molar basis, iodine is more cysticidal than hypochlorous acid.
2. Iodine has very little organic demand as compared to chlorine.
3. Chlorine has a strong affinity for nitrogenous matter, whereas iodine has almost none.
4. Both predominant forms of iodine, molecular iodine and hypoiodous acid, are efficient germicides. They form an excellent combination for cysts and enterovirus. On the other hand, where chlorine is used, hypochlorous acid alone is a good germicide.

Cysticidal Dose. Cysticidal doses of globaline and halazone tablets were determined in Cambridge tap water alone or with the addition of interfering substances that might be present in natural polluted water.[2] The cyst density employed, 60/ml of water, was considered to be far higher than the concentration of cysts in sewage. It was estimated that in an area of high endemicity, the ratio of amoebic cysts to *E. coli* would be of the order of 1 to 100,000. The number of coliform organisms discharged by an individual is estimated to be about 400 bil/day. An infected individual would discharge cysts in numbers varying from several hundred to some 10 mil/day. This ratio would make the number of cysts in concentrated sewage about 10/ml.

Table 4 indicates that one tablet of globaline should be able to disinfect all cysts, pathogenic bacteria, and spores. No conclusive tests were carried out against organisms of infectious hepatitis and other enteroviruses. However, from the work of Chang[4] it is now possible to speculate on the viricidal capacity of waters disinfected with globaline. At 10°C, the pH of tap water was lowered to 6.6 by the addition of one globaline tablet which left a residual of 6.9 ppm of iodine. At pH 6.6, about 5 per cent of the titrable iodine is in the form of HOI.[6] As a result, the hypoiodous acid concentration is about 0.35 ppm. This amount of HOI may not be sufficient to be viricidal. The high concentration of titrable iodine and the use of an acidic buffer result, therefore, in a high cysticidal but low viricidal efficiency.

Tests with halazone tablets showed that at room temperature about 5 tablets/qt of water were required to destroy all cysts in 10 min, whereas 2.5 tablets were effective in 30 min. In moderately to heavily polluted water at the same temperature, 7 tablets were needed for disinfection in 10 min and about 5 for disinfection in 30 min. Larger dosages of these tablets are required because:

1. The halazone tablet can release a maximum concentration of titrable chlorine equal to 2.3 ppm and HOCl equal to 1.1 ppm (as Cl_2). This is far less than the dose required under adverse conditions. Morris[3] reports that 10 ppm of HOCl are required for 99 per cent kill of *E. histolytica* in 10 min at 5°C.
2. The halazone tablet has an alkaline buffer to aid in dissolving the compound. Unfortunately, at high pH the predominant species of chlorine is OCl^- which is about 100 times less cysticidal than HOCl.
3. The dissolution time of the halazone tablet is slow; 7.5 minutes at room temperature.

On the other hand, the tablet has a great advantage over the globaline tablet in that HOCl is an excellent viricidal agent.[3,7] It appears safe to assume, therefore, that if a certain dose of HOCl is cysticidal, it is also sufficient for all types of enterovirus. From information available, it appears that several improvements may be possible in the preparation of halazone tablets. The first is the inclusion of an acidifying agent which will promote the formation of HOCl but which will not affect the solubility of the compound. The second is the substitution of a filler which will increase the rate of solution of the tablet.

Acceptability of Tablets by Users. The acceptance of the disinfecting agent by the user is probably as important as its germicidal action. The user may hesitate to use the agent because of unpleasant taste, odor or color; adverse physiological reaction; or excessive time for disinfection.

TABLE 4

Effectiveness of Globaline

Kind of Water	Temp °C	Contact Time *min*	pH		Cysticidal Dose *tablet/qt*	Cysticidal Residual *I₂-ppm*
			Initial	Final		
Tap	3	25	8.0 to 9.0	6.5	1	7.5
Tap	10	15	8.0 to 9.0	6.6	1	6.9
Tap	23	10	8.0 to 9.0	7.3	1	7.5
Tap	28	5	8.0 to 9.0	6.65	1	7.5
Tea infusion	23	5	7.2	6.4	2	8.7

1. Unpleasant Taste, Odor or Color.

Tastes and odors may be caused either by the tablet itself in water or by its combination with beverage powders. For purposes of comparison, the Whipple scale of intensity of odors and tastes[8] was adopted as a yardstick to determine the relative palatability of the tablets. Investigators[2] used 4 tablets of halazone providing about 10 ppm of titrable chlorine and 1 tablet of globaline providing 8 ppm of titrable iodine per liter of boiled distilled water at 23°C. The pH was varied with citric acid, dihydrogen disodium pyrophosphate, and sulfuric acid. The water was tested by seven to fourteen subjects. The results obtained indicate that in the "pH range commonly encountered," the globaline was more acceptable than halazone. In fact, in this range of pH, globaline produced "faint" to "distinct" intensity of odor and taste whereas halazone treated water was rated "decided" to "very strong" range on the Whipple scale. The "objectionable thresholds" were also determined in boiled distilled water at 23°C and the results are shown in Table 5.

It is apparent that globaline would reach the "objectionable threshold" only if 2 tablets were used as is prescribed for heavily polluted waters. As for the effect of pH upon tastes and odors, it was deduced, though not conclusively, that tastes and odors were minimal at the pH values attained when the tablets are added to neutral, unbuffered waters. To study the effect

TABLE 5

Per Cent of Normal Cysticidal Dose at Which "Objectionable Threshold" Is Reached

Compound	Percentage					
pH	4	5	6	7	8	9
Halazone	50	40	25	25	25	25
Globaline	—	200	—	200	—	—