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MECHANISM OF HARDNESS OF AGED COMPACTS

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ABSTRACT

The hardness changes in aged compacts of the chloride, bromide and iodide salts of sodium, potassium and ammonium, stored at various relative humidities, were studied. The effect of moisture on hardness followed a parabolic curve. Hardness increased with increasing moisture content, reached a maximum value in the region of deliquescence and then decreased drastically as the salt deliquesced. The incorporation of moisture in the starting material resulted in compacts with a pronounced decrease in strength. However, no further significant hardness changes were observed on aging. Addition of lubricants drastically reduced tablet hardness, and significantly reduced the hardening on aging.

INTRODUCTION

Physical aging of compressed tablets causes complex changes of hardness over a relatively short period. Hardness of compacts provides a direct measure of the

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bonding potential of a given solid material. The crushing strength of sodium chloride compacts prepared from dried samples doubled 1 hour after compaction[1]. No significant changes in crushing strength were observed when compacts were stored for longer periods at low humidities. Significant increases in hardness were observed in sodium chloride tablets stored for several days[2]. The effect of moisture on tablet hardness has recently been studied[3]. Significant hardness increase was measured for sodium, potassium, and ammonium chloride tablets stored at low humidities. Tablets stored at 76% relative humidity showed no increase or a slight decrease in hardness.

Therefore, the importance of aging and humidity on the strength of tablets has been recognized, but is still incomplete in many aspects and needs more clarification.

EXPERIMENTAL

Materials

Single lots of salts (Fisher Chemicals), magnesium stearate (Amend Drug) and polyethylene glycol 4000 (Ruger Chemicals) were selected for this investigation. Saturated solutions of salts were used to maintain various percent relative humidities in 5 liter desiccators.

Preparation and Treatment of Compacts

Batches of compacts were prepared from 30/40 mesh salt previously dried at 65°C for 24 hours. Tablets were compressed using a 1.27 cm. diameter die and punch assembly on a single punch machine (Manesty). Compacts of a suitable weight, thickness and hardness were produced and stored in open petri dishes at different relative humidities. The hardness of the compacts was determined at the time of making and at regular intervals thereafter. The construction and evaluation of the hardness tester used in this work has been described before[3].

Incorporation of Moisture

Moisture was incorporated by directing moist air onto the salt contained in a rotating 8 inch stainless steel coating pan. The spray nozzle of the plastic tube leading from a cool-mist (John Oster Manufacturing Company) humidifier was positioned near the opening of the coating pan at an angle so as to direct the moist air onto the material. The endpoint was arbitrarily selected when the material became visibly dampened. The humidified sample was then transferred to a suitable glass container with a screw type cap having an aluminum liner and stored for 24 hours with constant tumbling to assure homogeneity.

Incorporation of Lubricants

An approximate 250 g sample of 30/40 mesh salt was dried in a hot air oven at 65°C for 24 hours. The dried salt was blended with screened lubricant. Mixing was carried out in a V-type blender for fifteen minutes. The sample was then transferred into a dry glass bottle, sealed tightly with a screw type cap having an aluminum liner and stored over silica gel in a desiccator until needed.

RESULTS AND DISCUSSION

Hardness of Aged Compacts Stored at Different Humidities

Compacts of sodium, potassium and ammonium halides were prepared from dried materials and stored at several humidities. Tables 1-9 summarize the hardness determinations of these compacts at the time of making and at regular intervals thereafter. Figures 1 and 2 are a graphical representation of this data for the potassium compacts. The salts were stored at humidities above and below their critical deliquescent points. The critical deliquescent point is the relative humidity of air in equilibrium with the saturated solution of the salt since it is at this relative humidity that the substance should commence to deliquesce. Table 10 lists the literature values[4] for the critical deliquescent points of the salts.

TABLE 1

The Average[a] Hardness (kg) of Aged Compacts of Sodium Chloride Stored at Various Relative Humidities

Storage Period Days	20% (2.25[b]-2.27)	47% (2.29-2.295)	70% (2.37-2.47)	76% (2.29-2.30)	79% (2.37-2.48)
Initial	2.24 (0.22)	2.30 (0.25)	2.70 (0.17)	2.44 (0.21)	2.70 (0.17)
1	-	-	3.19 (0.38)	-	4.29 (0.07)
2	2.94 (0.30)	2.95 (0.35)	4.06 (0.40)	4.27 (0.25)	4.16 (0.17)
6	3.09 (0.21)	3.36 (0.28)	-	6.43 (0.39)	-
8	-	-	4.39 (0.30)	-	2.59 (0.12)
12	3.32 (0.34)	3.48 (0.34)	-	6.74 (0.35)	-
14	-	-	4.95 (0.20)	-	1.65 (0.12)
19	2.93 (0.30)	3.99 (0.44)	-	6.48 (0.52)	-
30	3.40 (0.09)	4.43 (0.40)	5.92 (0.20)	6.38 (0.12)	1.04 (0.11)

[a] Average of five observations

[b] Range of compact weight/thickness ratio
Standard deviation is given in parenthesis.

TABLE 2

The Average[a] Hardness (kg) of Aged Compacts of Sodium Bromide Stored at Various Relative Humidities

Storage Period Days	20% (3.28[b]-3.26)	47% (3.28-3.27)	52% (3.32-3.35)	66% (3.32-3.33)
Initial	5.88 (0.60)	5.88 (0.60)	3.37 (0.24)	3.37 (0.24)
1	5.36 (0.37)	7.36 (0.47)	4.98 (0.19)	4.64 (0.28)
3	-	-	-	4.77 (0.24)
4	5.40 (0.54)	8.20 (0.80)	4.95 (0.22)	-
6	4.68 (0.34)	9.62 (0.73)	4.61 (0.39)	-
7	-	-	-	4.88 (0.22)
26	5.68 (0.45)	9.30 (0.79)	4.86 (0.41)	*-

[a] Average of five observations

[b] Range of compact weight/thickness ratio

Standard deviation is given in parenthesis.

* Thereafter compacts deliquesced.

TABLE 3

The Average[a] Hardness (kg) of Aged Compacts of Sodium Iodide Stored at Various Relative Humidities

Storage Period Days	20% (3.47[b]-3.52)	32% (3.69-3.67)	43% (3.69-3.55)	47% (3.47-3.48)
Initial	2.04 (0.08)	3.18 (0.10)	3.18 (0.10)	2.04 (0.08)
3	3.74 (0.28)	4.77 (0.38)	4.12 (0.28)	3.68 (0.18)
7	4.42 (0.13)	4.96 (0.37)	3.77 (0.19)	*
13	5.26 (0.40)		*	

[a] Average of five observations

[b] Range of compact weight/thickness ratio

Standard deviation is given in parenthesis.

* Thereafter compacts deliquesced.

TABLE 4

The Average[a] Hardness (kg) of Aged Compacts of Potassium Chloride Stored at Various Relative Humidities

Storage Period Days	20% (2.19[b]-2.29)	47% (2.19-2.27)	76% (2.19-2.28)	79% (2.32-2.33)	90% (2.36-2.39)
Initial	1.87 (0.18)	1.87 (0.18)	1.87 (0.18)	2.72 (0.15)	3.52 (0.21)
1	-	-	-	5.32 (0.48)	3.29 (0.13)
3	2.58 (0.23)	3.19 (0.26)	6.38 (0.72)	5.53 (0.44)	2.43 (0.20)
7	2.71 (0.27)	3.89 (0.24)	7.64 (0.65)	5.85 (0.59)	2.24 (0.18)
12	2.98 (0.27)	4.06 (0.13)	10.17 (1.10)	-	-
16	-	-	-	6.07 (0.48)	2.00 (0.17)
20	3.74 (0.14)	4.57 (0.37)	9.01 (0.63)	-	-
30	3.69 (0.38)	4.32 (0.45)	10.67 (0.48)	6.45 (0.26)	1.68 (0.16)

[a] Average of five observations

[b] Range of compact weight/thickness ratio

Standard deviation is given in parenthesis.

TABLE 5

The Average[a] Hardness (kg) of Aged Compacts of Potassium Bromide Stored at Various Relative Humidities

Storage Period Days	20% (3.20[b]-3.22)	47% (3.20-3.27)	56% (2.33-2.35)	68% (2.33-2.37)	76% (3.20-3.23)	84% (2.34-2.26)
Initial	3.34 (0.22)	3.34 (0.22)	2.93 (0.21)	2.93 (0.21)	3.34 (0.02)	11.73 (1.18)
1	-	-	5.85 (0.40)	5.85 (0.40)	-	3.66 (0.29)
2	4.57 (0.45)	6.63 (0.39)	-	-	3.04 (0.31)	-
3	-	-	10.41 (0.25)	8.75 (0.56)	-	-
5	4.94 (0.36)	7.66 (0.62)	-	-	2.66 (0.24)	-
7	5.83 (0.43)	6.78 (0.60)	13.41 (0.73)	11.20 (0.93)	2.21 (0.12)	2.19 (0.10)
15	8.50 (0.52)	15.63 (0.62)	13.79 (0.70)	12.26 (0.36)	3.19 (0.23)	3.18 (0.06)
25	9.89 (1.19)	14.85 (2.01)	-	-	4.15 (0.50)	-
30	-	-	12.91 (1.91)	13.24 (0.67)	-	-
45	11.17 (1.20)	18.68 (1.84)	-	-	-	2.79 (0.38)

[a] Average of five observations

[b] Range of compact weight/thickness ratio

Standard deviation is given in parenthesis.

TABLE 6

The Average[a] Hardness (kg) of Aged Compacts of Potassium Iodide Stored at Various Relative Humidities

Storage Period Days	20% (3.56[b]-3.57)	47% (3.56-3.56)	56% (3.61-3.60)	70% (3.61-3.60)	76% (3.56-3.61)
Initial	5.72 (0.54)	5.72 (0.54)	2.78 (0.23)	2.78 (0.23)	5.72 (0.54)
1	7.29 (0.50)	9.17 (0.95)	3.81 (0.39)	0.79 (0.00)	0.89 (0.07)
3	6.96 (0.58)	10.49 (1.03)	2.44 (0.13)	1.13 (0.13)	1.70 (0.20)
7	6.73 (0.52)	8.30 (0.88)	2.38 (0.28)	0.62 (0.05)	1.55 (0.15)
14	-	-	2.08 (0.20)	0.35 (0.03)	-
27	6.49 (0.16)	9.78 (1.16)	-	-	1.80 (0.18)
31	-	-	2.11 (0.10)	0.28 (0.02)	-

[a] Average of five observations

[b] Range of compact weight/thickness ratio

Standard deviation is given in parenthesis.

TABLE 7

The Average[a] Hardness (kg) of Aged Compacts of Ammonium Chloride Stored at Various Relative Humidities

Storage Period Days	20% (1.68[b]-1.75)	47% (1.74-1.76)	76% (1.74-1.78)	84% (1.69-1.74)
Initial	1.86 (0.17)	1.83 (0.12)	1.83 (0.12)	2.02 (0.10)
1	-	-	-	1.72 (0.09)
3	1.95 (0.19)	2.49 (0.12)	4.01 (0.36)	1.57 (0.07)
7	2.28 (0.15)	2.86 (0.27)	3.33 (0.13)	-
12	3.07 (0.14)	2.67 (0.27)	3.32 (0.16)	-
15	-	-	-	1.13 (0.07)
21	3.33 (0.27)	3.25 (0.33)	3.12 (0.15)	-
30	3.39 (0.18)	3.57 (0.11)	2.89 (0.18)	-

[a] Average of five observations

[b] Range of compact weight/thickness ratio

Standard deviation is given in parenthesis.

TABLE 8

The Average[a] Hardness (kg) of Aged Compacts of Ammonium Bromide Stored at Various Relative Humidities

Storage Period Days	20% (2.77[b]-2.86)	47% (2.78-2.84)	76% (2.77-2.79)	79% (2.80-2.97)	90% (2.80-2.86)
Initial	2.50 (0.12)	-	2.50 (0.12)	2.41 (0.10)	2.41 (0.10)
1	2.40 (0.27)	2.77 (0.29)	4.79 (0.43)	1.08 (0.12)	0.96 (0.05)
3	2.12 (0.30)	2.77 (0.16)	4.11 (0.39)	1.12 (0.04)	0.91 (0.02)
7	2.90 (0.24)	2.71 (0.073)	3.29 (0.34)	1.02 (0.05)	-
15	-	-	-	0.94 (0.05)	-
25	3.22 (0.44)	2.72 (0.16)	2.20 (0.16)	-	-

[a] Average of five observations

[b] Range of compact weight/thickness ratio

Standard deviation is given in parenthesis.

TABLE 9

The Average[a] Hardness (kg) of Aged Compacts of Ammonium Iodide Stored at Various Relative Humidities

Storage Period Days	20% (2.78[b]-2.77)	47% (2.78-2.71)	56% (2.89-2.82)	70% (2.89-2.90)	76% (2.78-2.81)
Initial	5.89 (0.29)	5.89 (0.29)	2.08 (0.17)	2.08 (0.17)	5.89 (0.29)
1	11.73 (1.34)	12.5 (0.46,	0.46 (0.00)	0.15 (0.01)	0.65 (0.06)
3	10.84 (0.95)	14.87 (0.49)	0.54 (0.03)	0.20 (0.02)	0.51 (0.07)
8	-	-	0.50 (0.03)	-	-
16	-	-	0.60 (0.05)	-	-
24	13.31 (0.54)	*	-	-	0.40 (0.06)
30	-	-	0.72 (0.08)	-	-

[a] Average of five observations

[b] Range of compact weight/thickness ratio

Standard deviation is given in parenthesis.

* Compacts deliquesced.

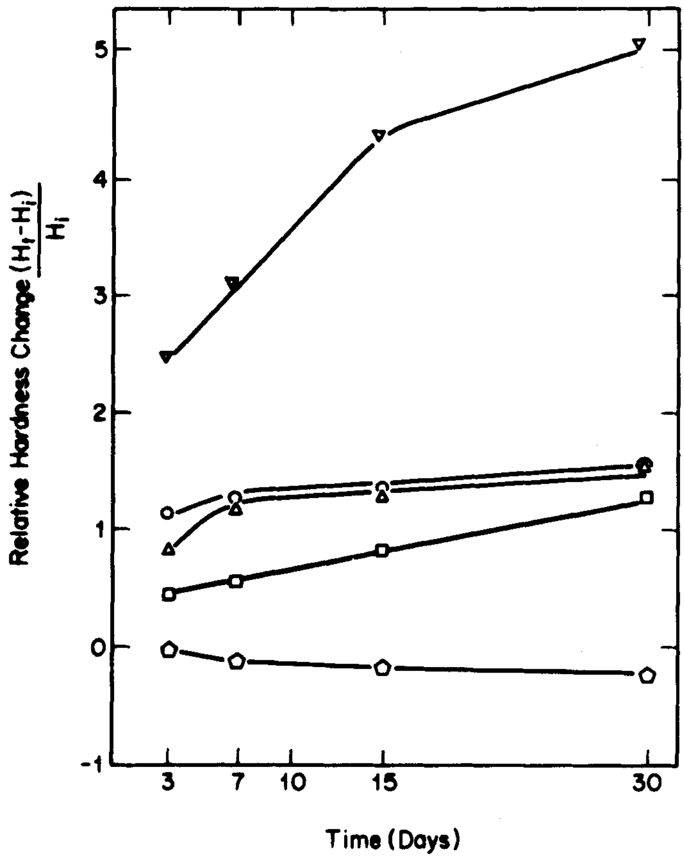

FIGURE 1

Relative Hardness Changes Observed in Potassium Chloride Compacts Stored at Different Relative Humidities

(H_t - hardness at time t

H_i - initial hardness)

Key: □ - 20%

△ - 47%

▽ - 76%

○ - 79%

⬠ - 90%

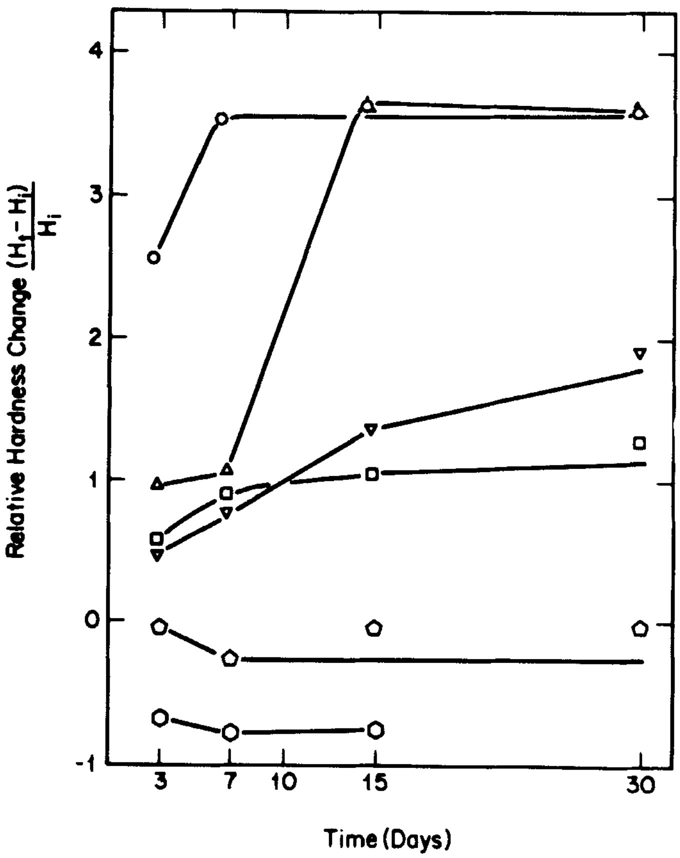

FIGURE 2

Relative Hardness Changes Observed in Potassium Bromide Compacts Stored at Different Relative Humidities

(H_t - hardness at time t

H_i - initial hardness)

Key: ▽ - 20%
△ - 47%
○ - 56%
□ - 68%
⬠ - 76%
⬡ - 84%

TABLE 10

Critical Deliquescent Points for the Salts at 25°

Salt	Critical Deliquescent Point (% Relative Humidity)
Sodium Chloride	75.3
Sodium Bromide	56.0
Sodium Iodide	38.7
Potassium Chloride	84.3
Potassium Bromide	80.9
Potassium Iodide	68.9
Ammonium Chloride	79.3
Ammonium Bromide	76.0
Ammonium Iodide	55.0

The small strength increases observed at 20% humidity are due to time-dependent relaxation which provides stress relief within the compact. Increased bonding may occur if elastic or plastic flow increases the area of intimate contact between adjacent crystals[1].

As the humidity increases but remains below the critical deliquescent point, the hardness of the salts increases. At a particular humidity, hardness would probably change until the compact attains its equilibrium moisture content. Thereafter the hardness would remain constant. This may also indicate that all sites for water-substrate interaction are saturated.

Shotton et al.[1] observed increases in strength of sodium chloride compacts stored at 66% relative humidity and suggested that this increase may have been caused by dissolution and removal of surface cracks and flaws in the crystals, which otherwise act as centers of stress concentration. In studies of granular salts, Hall and Rose[5] found that the ammonium chloride and iodide salts caked with increasing moisture content and suggested that the cohesive force involved in the caking process is capillary adhesion. They state that capillary adhesion occurs when adsorbed water condenses around the particles and coalesces in the gap between the two bodies. The meniscus so formed draws on the particles by means of surface tension. These authors did not measure the strength of the cakes but it is very likely that the strength would have increased with caking. Thompson[6] has suggested a caking mechanism for ammonium nitrate which involves plastic deformation. He states that with increasing moisture contents its plasticity increases, allowing areas of close contact to be formed between particles.

Above the critical deliquescent point, the hardness of the salts decreased considerably. Shotton et al.[1] found that the crushing strength of sodium chloride compacts stored at 76% relative humidity for eight weeks decreased considerably. They attribute this effect to be due to extensive dissolution and subsequent recrystallization which could occur repeatedly following small fluctuations in the storage conditions. Consequently, they state that the compact no longer represents the original compacted system, since the interparticulate bonds formed during compaction are removed by dissolution. New bonds then form by recrystallization of dissolved solid material within the compact. The increased number of discontinuities which can propagate failure of the compact contributes appreciably to the pronounced decrease in strength. The decrease in hardness may also be due to the significant increase in moisture content of the compact which would separate the crystals from each other and thereby minimize the effect of the inherent forces of cohesion.

Therefore, moisture picked up by storage at humidities below the critical deliquescent point helps heal any fractures or flaws present in the internal surface of the compact. Thus, points of contact are brought closer to each other and bond formation is facilitated with a resultant increase in compact hardness.

With an increase in moisture uptake, brought about by storage at humidities near or above the critical deliquescent point, some interparticulate bonds are probably dissolved. Accordingly, the points of contact are separated from each other resulting in a decreased magnitude of the molecular forces of attraction. This is manifested in a considerable reduction in compact strength.

Additional samples of potassium chloride and bromide compacts exposed to humid conditions were dried at 105°C for 2 hours and cooled in a desiccator before their hardness was determined. The effects of humidity and treatment are shown in Table 11.

The data suggests that drying of the compacts did not appreciably affect their strength. This observation supports the earlier conclusions that the principal mechanism of hardening occurs because of moisture picked up by the

TABLE 11

The Hardness of Potassium Chloride and Potassium Bromide Compacts Stored at Elevated Humidities for Two Weeks

		Tensile Strength - kg	
Storage Conditions	Treatment	Potassium Chloride	Potassium Bromide
Initial	-	1.72	2.82
47% RH	-	7.42	7.07
47% RH	Dried	7.45	9.14
76% RH	-	8.69	2.31
76%	Dried	8.74	2.99

salts on storage at humidities below their critical deliquescent point and not because of any loss of moisture.

The relative hardness changes observed in chloride salts of sodium, potassium, and ammonium stored at 47% relative humidity, is shown in Figure 3. In all instances, moisture assists bond formation. Potassium chloride has the highest critical deliquescent point and experiences a greater hardness increase

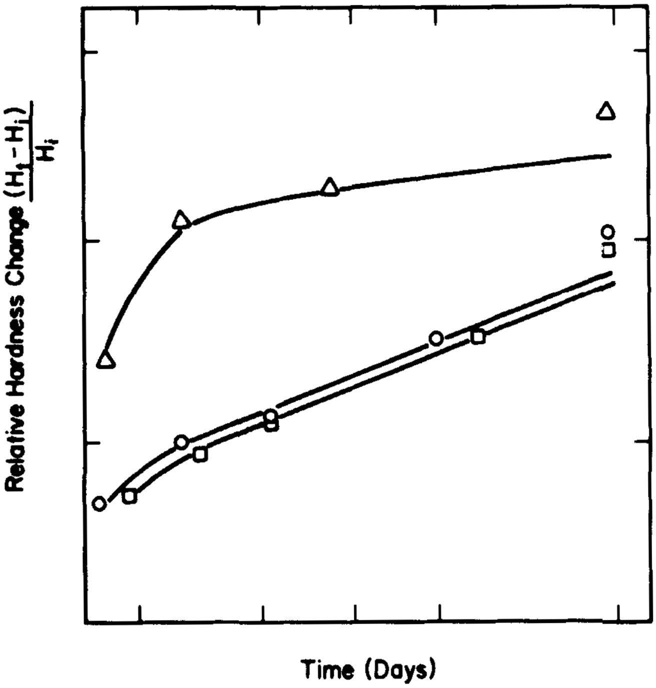

FIGURE 3

Relative Hardness Changes Observed in Compacts of Chloride Salts Stored at 47% Relative Humidity

(H_t - hardness at time t

H_i - initial hardness)

Key:

○ - Sodium Chloride

△ - Potassium Chloride

□ - Ammonium Chloride

with time than the other two salts. The values of the critical deliquescent points for the sodium and ammonium salts are close to each other. This is reflected in their similar tendency towards hardening with time.

The relative hardness changes in potassium salts stored at 47% relative humidity is shown in Figure 4. The effect of moisture on potassium iodide compacts, which have the lowest critical deliquescent point, is to disrupt bond

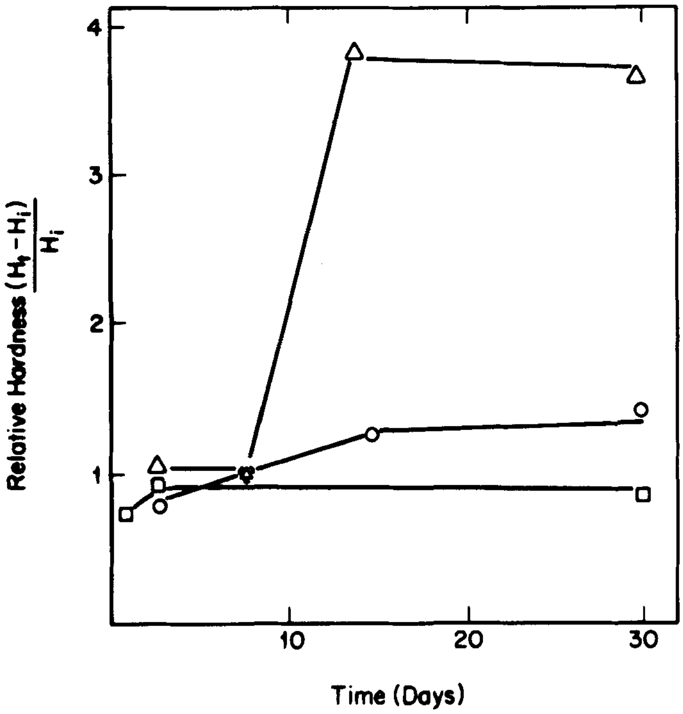

FIGURE 4

Relative Hardness Changes Observed in Compacts of Potassium Salts Stored at 47% Relative Humidity

(H_t - hardness at time t

H_i - initial hardness)

Key:

○ - Potassium Chloride

△ - Potassium Bromide

□ - Potassium Iodide

formation. This is reflected in the gradual reduction in hardness of compacts with time. Moisture assists bond formation to the largest extent for potassium bromide which has an intermediate value for the critical deliquescent point - 80%. Moisture also assists bond formation in the potassium chloride compacts, which have the highest critical deliquescent point - 84%. However, this effect is not as pronounced as for the potassium bromide salt.

1. Effect of Pre-Treatment

a. Non-drying of the Material

Compacts of potassium bromide were prepared from the salt which had not been dried prior to compression. The compacts were then stored at 47%, 56%, 68% relative humidity. The hardness values are summarized in Table 12. Measurements were made on compacts of equal density, as indicated by the

TABLE 12

The Hardness [a]kg of Potassium Bromide Compacts
Stored at Various Relative Humidities at 20°C

Storage Period Days	Relative Humidity					
	47%		56%		68%	
Initial	2.65	(0.17)	-		-	
1	3.81	(0.16)	3.71	(0.11)	3.01	(0.08)
3	5.01	(0.00)	5.01	(0.24)	3.33	(0.16)
7	6.39	(0.62)	5.66	(0.45)	3.29	(0.29)
15	7.25	(0.70)	5.77	(0.49)	3.06	(0.20)

[a] Average of five observations

Standard deviation is given in parenthesis.

constancy of the weight to thickness ratio. The data suggests that the mechanism of hardening occurs because of a loss of traces of moisture at low humidities. The loss of moisture results in the deposition of salt around existing contact regions which acts as a solid bridge. The extent of moisture loss is greatest at 47% relative humidity which is reflected in the greater strength of the compacts stored at this humidity. Additional data to support the above conclusion was obtained by preparing potassium bromide compacts from undried starting material. These compacts were stored at 47% relative humidity and 47°C. Table 13 summarizes the hardness values measured over a period of time. The increase in hardness is due to loss of moisture and formation of solid bridges by the recrystallization of the dissolved salt in the voids created.

b. Incorporation of Moisture in the Material

Compacts of potassium chloride and bromide salts were prepared from the materials within which moisture had been introduced. The compacts were then stored at 20% and 76% relative humidities. The hardness values are presented in Table 14. Incorporation of moisture into the starting material considerably decreased the strength of the compacts and inhibited significant hardness changes in aging.

2. Effect of Lubricant

In order to study the effect of hydrophilic and a hydrophobic lubricant on the hardness of aged salts, compacts of potassium chloride blended with 0.3% polyethylene glycol and 0.5% stearic acid, respectively, were prepared. These compacts were stored at different humidities. Table 15 shows the hardness values. Measurements were made on compacts of equal density, as indicated by the constancy of the weight to thickness ratio.

The lubricants coat the salt crystals forming an insulating layer and thereby minimizing the inherent molecular forces of attraction. This is reflected in the

TABLE 13

The Hardness [a]kg of Aged Compacts of Potassium Bromide

Storage Period Days	47% RH	47°C
Initial	2.82 (0.22)	-
2	5.34 (0.37)	6.59 (0.37)
5	6.43 (0.43)	6.59 (0.62)
11	6.92 (0.61)	7.05 (0.70)
20	9.14 (0.84)	8.34 (0.58)

[a] Average of five observations
Standard deviation is given in parenthesis.

TABLE 14

The Hardness [a]kg of Aged Compacts of Salts Stored at Various Relative Humidities

Storage Period Days	Potassium Chloride 20%	Potassium Chloride 76%	Potassium Bromide 20%	Potassium Bromide 76%
Initial	1.84 (0.14)	-	1.65 (0.16)	-
9	2.99 (0.28)	2.12 (0.28)	2.14 (0.20)	1.70 (0.18)
13	2.29 (0.18)	2.33 (0.25)	2.16 (0.14)	1.76 (0.13)
21	3.23 (0.32)	2.58 (0.31)	2.58 (0.17)	1.54 (0.03)
31	2.76 (0.15)	2.49 (0.25)	2.36 (0.30)	1.49 (0.12)

[a] Average of five observations

Standard deviation is given in parenthesis.

TABLE 15

The Hardness [a]kg of Aged Compacts of Potassium Chloride Salts Stored at Various Relative Humidities

Storage Period Days	With Polyethylene Glycol 20%	With Polyethylene Glycol 47%	With Polyethylene Glycol 76%	With Stearic Acid 20%	With Stearic Acid 47%	With Stearic Acid 76%
Initial	0.97 (0.07)	-	-	0.47 (0.03)	0.49 (0.01)	0.49 (0.05)
3	2.00 (0.24)	2.33 (0.04)	1.63 (0.18)	0.56 (0.03)	0.51 (0.02)	0.60 (0.04)
15	1.99 (0.09)	1.90 (0.10)	2.00 (0.06)	0.53 (0.02)	0.48 (0.04)	0.63 (0.03)
30	1.96 (0.07)	2.04 (0.07)	2.07 (0.09)	0.57 (0.05)	0.62 (0.06)	0.66 (0.02)

[a] Average of five observations

Standard deviation is given in parenthesis.

considerable decrease in the strength of the compacts. The magnitude of this effect is less when the hydrophilic lubricant, polyethylene glycol 4000, is employed. This may be attributed to the thread-like binding property of the polymer. In both cases there is an initial hardness increase followed by stabilization where neither aging nor humidity exerts any effect on the strength. The initial hardness increase may be attributed to the stress relaxation phenomenon discussed in an earlier section.

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