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A THERMODYNAMIC STUDY OF THE AC-DI-SOL-WATER AND CLD-2-WATER SURFACE INTERACTION

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ABSTRACT

Adsorption-desorption studies and immersional calorimetry were used conjointly to characterize the surface properties of two disintegrants - Ac-Di-Sol and CLD-2. Adsorption-desorption isotherms were utilized to calculate the integral and different Gibb's free energies. Non-isothermal calorimetry was employed to quantify the energies of interaction between the adsorbate and the adsorbent and obtain the differential enthalpy. These interactions were determined to be -7.5 and -7.8 kcal per mole for Ac-Di-Sol and CLD-2, respectively. The premature decrease in the differential enthalpies and entropies and the observed immersional and adsorption-desorption hystereses were hypothesized to be the result of an irreversible swelling

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process. Differences were observed between CLD-2 and Ac-Di-Sol which were believed to be due to the higher degree of cross-linking in Ac-Di-Sol.

INTRODUCTION

The interaction of water with pharmaceutical excipients is usually of considerable importance to the ultimate release of the drug from the dosage form. This is particularly true for the water-disintegrant interaction. Recently, the water sorption properties of a number of newer disintegrating agents have been investigated[1,2]. In the present investigation, a more complete characterization of Ac-Di-Sol and CLD-2 has been achieved using aqueous immersional calorimetry.

EXPERIMENTAL

Water Vapor Sorption

The water vapor adsorption-desorption studies were necessary to characterize thermodynamically the integral and differential Gibb's free energies for the adsorption process of water vapor on to Ac-Di-Sol and CLD-2. Ac-Di-Sol and CLD-2, approximately 60 to 80 grams, were initially vacuum dried (0.1 mm Hg) at 60°C for five days to remove all the adsorbed gases from their respective surfaces. Each of the dried disintegrants, approximately 1 to 3 grams, were placed in previously weighed petri dish tops and transferred to relative humidity chambers ranging from 7 to 100% relative humidity. These chambers were prepared by filling Pyrex desiccators with a saturated salt solution which would maintain the desired relative humidity at 25°C. The chambers were stored in an environmental control room maintained at 25°C ± 0.05°C.

The adsorption of water vapor by the dried disintegrants was followed gravimetrically as a function of time; weighings were performed approximately every 48 hours. The gravimetric determination of the change in weight resulting from the adsorption or desorption processes was determined by difference using a Mettler balance with a sensitivity of 0.1 mg.

The desorption studies were performed in a manner similar to the adsorption studies with the exception that the dried disintegrants were first equilibrated at 100% relative humidity before being transferred to various relative humidity chambers. Weight determinations were made approximately every 48 hours.

The reversibility of the adsorption process for both disintegrants was investigated by combining an adsorption, desorption and re-adsorption sequence into one study. The dried disintegrants were initially equilibrated at 100% relative humidity (adsorption process), then dried at 60°C in the vacuum oven for five days (desorption process), and finally re-equilibrated in various relative humidity chambers for seven days (re-adsorption process). The total net change in weight incurred during the 7 day re-adsorption phase was compared with the total weight change of those samples which had undergone the adsorption process only once.

Aqueous Immersional Calorimetry

Heats of immersion of Ac-Di-Sol and CLD-2 containing various amounts of adsorbed water and water remaining after the desorption process were determined in a nonisothermal calorimeter. The design of this calorimetry and aspects of its operation were presented previously[3].

A thin-walled glass sample bulb was tared and partially filled with powder equilibrated under one of the previously described conditions. The weight of the sample was obtained by the difference between the filled and empty sample bulbs.

The calorimeter cell was filled with an accurately measured quantity of distilled water (either 200 or 250 ml), and the entire unit was assembled. Stirring was initiated, and the temperature of the calorimeter contents, θ, was adjusted to approximately 25° by actuation of the calibration heater.

Thermal equilibration of the system was assumed complete when the observed $\Delta\theta / \Delta t$ values were not greater that $\pm$ 0.0002°/interval. (The digital thermometer reports the temperature at 10-sec intervals when operated at a resolution of 0.0001°). After stabilization, a preprocess period of at least 5 min (30 intervals) was obtained. The sample bulb was then fractured by a rapid depression and release of the beaker rod.

When the wetting process was complete as indicated by a temperature change per interval approaching those observed

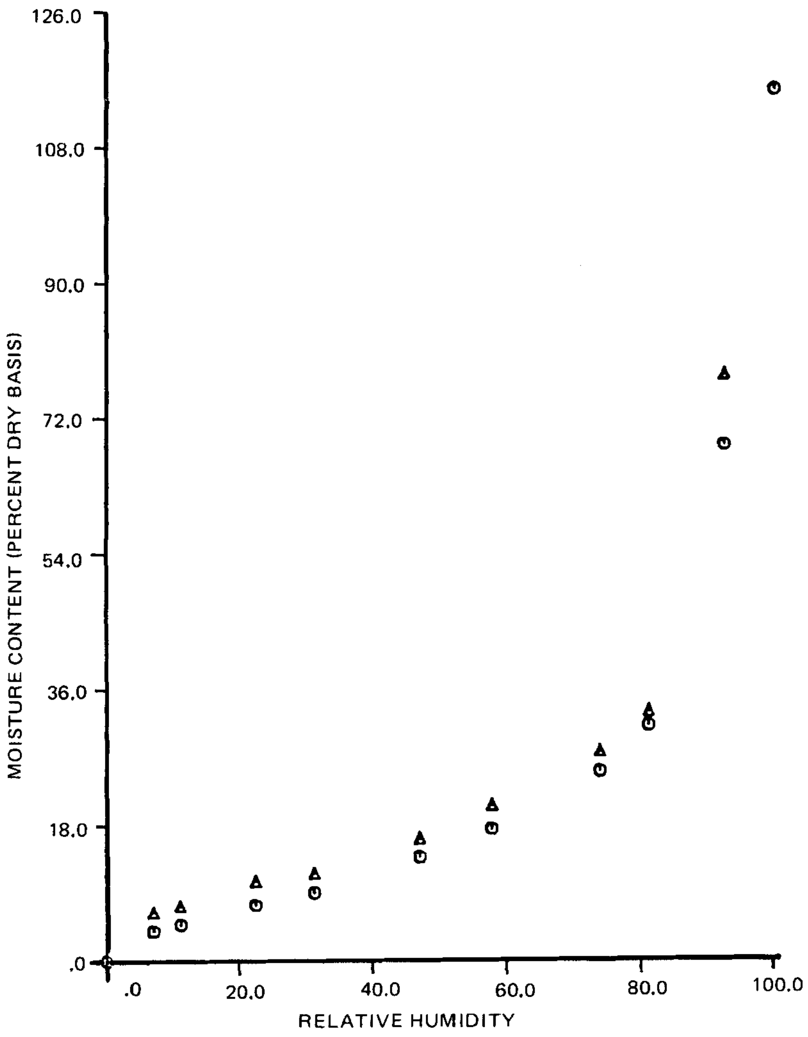

FIGURE 1. Ac-Di-Sol

during the initial preprocess period, a calibration was accomplished by actuation of the resistance heater in the calorimeter cell for approximately 10 sec, during which the voltage across the heater was measured. After termination of the heating process, the duration of the actuation was recorded.

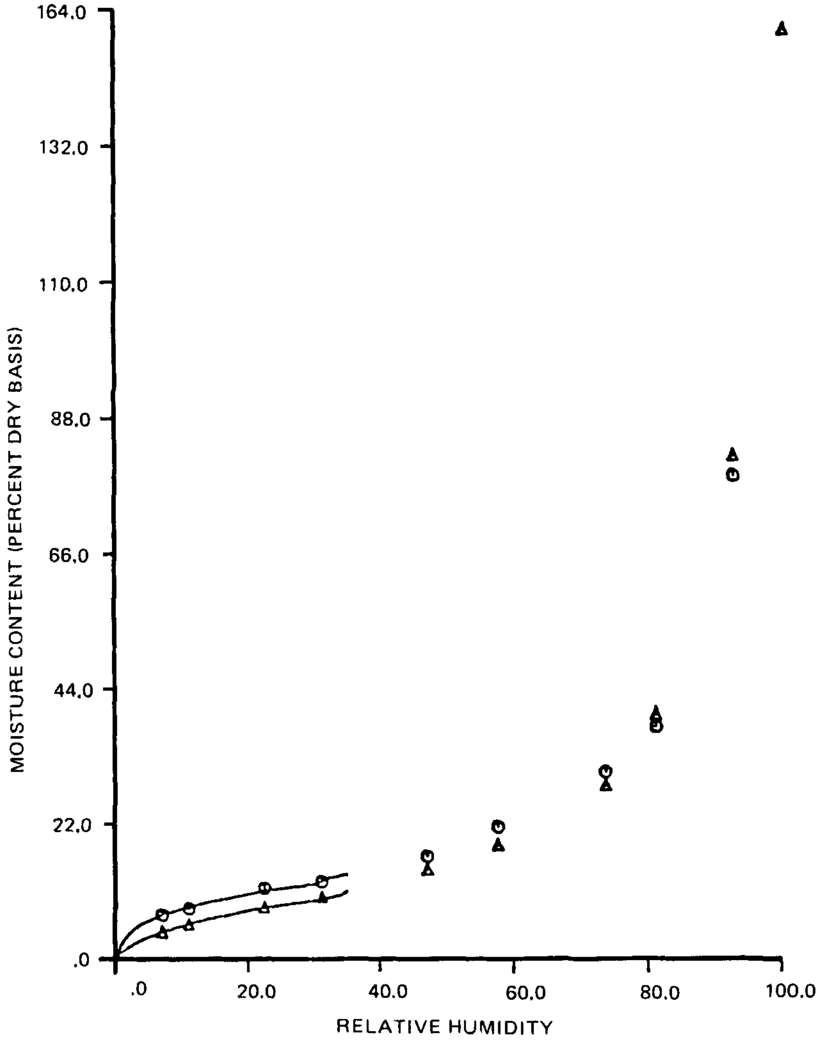

FIGURE 2. CLD 2

A 5-minute post actuation period was recorded before termination of the experiment. This procedure was employed for all samples.

Intrinsic temperature changes for the wetting and calibration processes were computed using the differential transformation method described previously[3,4]. The heat capacity used to compute the heats of wetting was obtained by averaging the results of all calibrations within the same set of experiments. The accuracy of all heats of wetting is related to the accuracy of the heat capacity determination for the system which was approximately ± 0.6%.

RESULTS AND DISCUSSION

The integral and differential Gibb's free energy changes associated with the adsorption of water by Ac-Di-Sol and CLD-2 were calculated from moisture content and relative humidity (partial pressure) values taken at selected intervals from the water vapor adsorption isotherms (Figures 1 and 2). The integral Gibb's free energy change was then calculated by the following equation:

$$\Delta G = -RT \int_{o}^{P/Po} nd(\ln \frac{Po}{P}) - nRT \ln P/Po \qquad (1)$$

Evaluation of this equation is complicated, however, by the fact that data at low pressures, which are seldom available, are required for quantification of the integral term. Also of major concern was the required extrapolation of the n vs. ln P/Po curve to zero pressure which is tedious and subject to large errors. However, an equivalent form of the integral term which substantially reduces the error incurred is

$$\Delta G = -RT \int_{o}^{P/Po} n \left(\frac{Po}{P} \right) d \left(\frac{P}{P_o} \right) - nRT \ln \frac{Po}{P} \qquad (2)$$

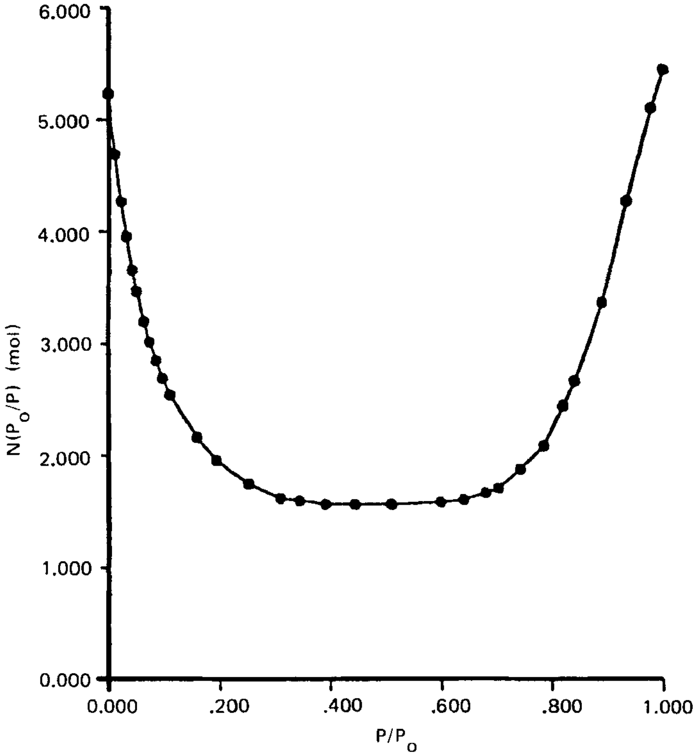

FIGURE 3. Modified Adsorption Isoltherm for Ac-Di-Sol-Water System

Construction of the n (Po/P) vs. P/Po graph can be easily extrapolated to zero pressure using linear extrapolation, as illustrated in Figures 3 and 4.

The integral can be evaluated by determining the area under the curve, which was accomplished using the trapezoidal rule. The results of this method are displayed in column three of Tables 1 and 2.

The differential Gibb's free energy change was calculated from

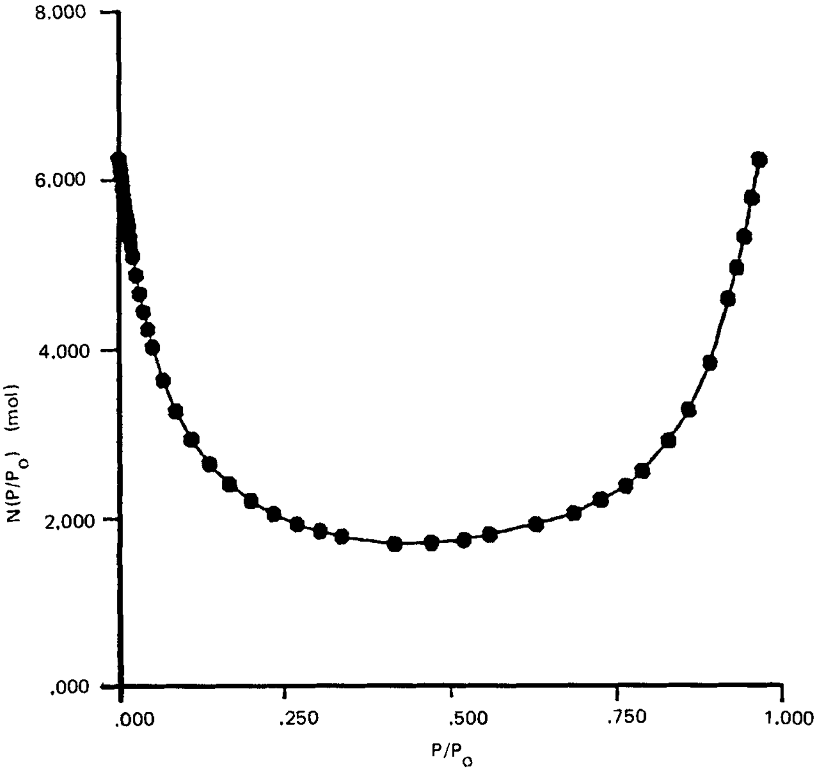

FIGURE 4. Modified Adsorption Isotherm for CLD-2-Water System.

$$\overline{\Delta G} = RT \ln {}^{P}/Po \qquad (3)$$

The enthalpy changes (H_L), as calculated from the heats of wetting[3,4], were obtained for both disintegrants containing various amounts of water vapor following attainment of equilibrium from the adsortion and desorption processes. These data are presented in Table 3 and 4 and illustrated in Figures 5 and 6.

TABLE 1

Thermodynamic Values for the Adsorption of Water by Ac-Di-Sol at 25° C.

n^a	$\frac{P}{P_o}$	$-RT\int_o^x \frac{nP_o}{P} d(\frac{P}{P_o})^b$	$-\Delta G^b$	$-\Delta H^b$	$-\Delta S^b$	$-\overline{\Delta G}^b$	$-\overline{\Delta H}^b$	$-\overline{\Delta S}^a$
0.01	0.001(9)	6	43	60	0.06	3697	6010	7.76
0.02	0.004(0)	12	78	119	0.14	3273	5950	8.98
0.03	0.006(1)	18	109	179	0.23	3019	5950	9.80
0.04	0.008(3)	25	138	238	0.34	2836	5950	10.41
0.05	0.010(6)	31	165	297	0.44	2690	5950	10.90
0.06	0.013(1)	38	192	357	0.55	2569	5950	11.31
0.07	0.015(6)	45	217	416	0.67	2464	5950	11.69
0.08	0.018(3)	52	241	476	0.79	2370	5950	11.97
0.09	0.021(1)	59	265	536	0.91	2286	5950	12.25
0.10	0.024(0)	65	287	595	1.03	2210	5950	12.51
0.11	0.027	74	309	654	1.16	2138	5950	12.75
0.12	0.030	81	330	714	1.29	2072	5950	13.01
0.13	0.033	89	350	773	1.42	2009	5950	13.18
0.14	0.037	97	370	833	1.55	1950	5950	13.38
0.15	0.041	105	389	892	1.69	1894	5950	13.57
0.16	0.045	114	408	951	1.82	1839	5950	13.75
0.17	0.049	122	426	1011	1.96	1788	5950	13.96
0.18	0.053	131	444	1070	2.10	1738	5950	14.09
0.19	0.058	140	461	1130	2.24	1689	5950	14.26
0.20	0.063	149	478	1189	2.39	1642	5950	14.41
0.22	0.073	168	510	1308	2.68	1552	5950	14.73
0.24	0.084	188	540	1427	2.98	1467	5950	15.00
0.26	0.097	208	568	1546	3.28	1385	5950	15.28
0.28	0.110	229	595	1665	3.59	1307	5950	15.55
0.30	0.125	251	626	1782	3.90	1233	5895	15.64

0.32	0.141	273	645	1890	4.17	1162	5380	14.15
0.34	0.157	295	667	1980	4.40	1095	4500	11.42
0.36	0.175	317	688	2050	4.57	1032	3500	8.28
0.38	0.194	339	708	2115	4.72	973	3250	7.64
0.40	0.213	360	727	2180	4.87	917	3250	7.82
0.42	0.232	382	745	2240	5.01	866	3000	7.16
0.44	0.251	402	762	2296	5.14	818	2800	6.65
0.46	0.271	422	778	2350	5.27	774	2700	6.45
0.48	0.290	441	793	2400	5.39	733	2500	5.93
0.50	0.309	459	807	2448	5.50	696	2400	5.72
0.55	0.344	493	840	2550	5.74	632	2040	4.72
0.60	0.392	537	870	2660	6.00	555	2200	5.52
0.70	0.445	586	922	2848	6.46	480	2100	5.43
0.80	0.510	646	965	3013	6.87	399	1600	4.03
0.90	0.580	713	1001	3137	7.16	305	1240	3.09
1.00	0.620	748	1031	3253	7.45	283	1100	2.74
1.20	0.702	829	1080	3463	7.99	210	1050	2.82
1.40	0.743	872	1119	3598	8.31	176	675	1.67
1.60	0.786	923	1151	3676	8.47	142	388	0.82

[a] $\frac{\text{moles adsorbate}}{\text{100 gm solid}}$.

[b] cal/100 gm.

[c] e.u./100 gm.

TABLE 2

Calculated Thermodynamic Properties for the Adsorption of Water by CLD-2 at 25° C.

n^a	$\frac{P}{P_o}$	$-RT\int_o^x \frac{nP_o d(\frac{P}{P_o})}{P}^b$	$-\Delta G^b$	$-\Delta H^b$	$-\Delta S^b$	$-\overline{\Delta G}^b$	$-\overline{\Delta H}^b$	$-\overline{\Delta S}^a$
0.01	0.001(6)	6	44	66	0.07	3803	6620	9.41
0.02	0.003(3)	12	80	132	0.18	3381	6620	10.83
0.03	0.005(1)	18	112	198	0.29	3129	6620	11.71
0.04	0.006(9)	25	142	265	0.41	2947	6620	12.28
0.05	0.008(8)	31	171	331	0.53	2803	6620	12.77
0.06	0.010(8)	38	199	397	0.66	2683	6620	13.20
0.07	0.012(8)	44	225	463	0.80	2579	6620	13.52
0.08	0.015(0)	51	250	529	0.93	2488	6620	13.83
0.09	0.017(3)	58	275	595	1.07	2405	6620	14.14
0.10	0.019	65	298	661	1.22	2330	6620	14.36
0.11	0.022	73	321	727	1.36	2260	6620	14.59
0.12	0.025	80	344	794	1.51	2196	6620	14.84
0.13	0.027	88	365	860	1.66	2135	6620	15.01
0.14	0.030	96	386	926	1.81	2077	6620	15.20
0.15	0.033	104	407	992	1.96	2022	6620	15.42
0.16	0.036	112	427	1058	2.12	1969	6620	15.56
0.17	0.039	120	446	1124	2.27	1920	6620	15.73
0.18	0.043	128	465	1190	2.43	1871	6620	15.93
0.19	0.046	137	484	1257	2.59	1824	6620	16.05
0.20	0.050	146	502	1323	2.75	1779	6620	16.24
0.22	0.057	164	536	1455	3.08	1692	6620	16.51
0.24	0.066	183	569	1587	3.41	1610	6620	16.77
0.26	0.075	203	600	1720	3.75	1531	6620	17.05
0.28	0.086	223	631	1852	4.09	1456	6620	17.29
0.30	0.097	244	659	1984	4.44	1383	6620	17.55

0.32	0.109	266	686	2116	4.80	1313	6620	17.78
0.34	0.122	288	712	2249	5.15	1246	6620	18.01
0.38	0.151	333	759	2325	5.25	1120	3500	7.98
0.40	0.166	356	780	2390	5.40	1062	3250	7.34
0.42	0.183	379	801	2450	5.53	1006	3150	7.19
0.44	0.199	401	822	2505	5.65	954	3750	9.38
0.46	0.217	424	839	2555	5.75	905	3500	8.70
0.48	0.235	445	858	2605	5.86	859	3500	8.80
0.50	0.252	466	874	2665	6.01	816	3000	7.32
0.54	0.287	506	905	2777	6.28	739	2760	6.77
0.60	0.337	560	947	2933	6.66	644	2500	6.22
0.70	0.415	640	1005	3180	7.30	521	2200	5.63
0.80	0.471	696	1053	3400	7.87	446	2100	5.55
0.90	0.520	746	1095	3595	8.39	387	1900	5.07
1.00	0.558	785	1132	3760	8.82	346	1500	3.87
1.10	0.595	826	1164	3880	9.11	308	1200	2.99
1.20	0.628	862	1193	3970	9.31	276	900	2.09
1.40	0.685	929	1243	4095	9.56	224	550	1.09
1.60	0.726	981	1284	4175	9.70	190	300	0.36
1.80	0.763	1030	1319	4235	9.78	160	250	0.29
2.0	0.788	1066	1349	4280	9.83	142	200	0.20
2.5	0.845	1161	1421	4355	9.84	100	100	0.00
3.0	0.8725	1214	1457	4375	9.79	81	25	-0.19
4.0	0.911	1305	1523	4391	9.61	55	9	-0.15
5.0	0.942	1390	1569	4401	9.53	36	15	-0.07
6.0	0.964	1467	1598	4410	9.43	22	0.0	-0.73

[a] moles adsorbate/100 gm solid.

[b] cal/100 gm.

[c] e.u./100 gm.

TABLE 3

Heats of Wetting of Ac-Di-Sol, CLD-2, and Amberlite IRP-88 Containing Various Amounts of Water for the Adsorption and Desorption Processes.

Ac-Di-Sol			
Adsorption		Desorption	
Moisture content (gm H_2O/100 gm solid)	$-\Delta H_i$(SFL) (cal/gm solid)	Moisture content (gm H_2O/100 gm solid)	$-\Delta H_i$(SFL) (cal/gm solid)
0.0	37.1	6.5	17.7
3.7	24.9	6.6	17.2
3.7	24.8	7.5	15.0
4.6	21.8	8.0	14.1
4.8	21.1	11.0	10.3
8.0	15.4	11.4	9.7
8.9	13.6	11.7	9.5
9.1	13.6	12.7	8.1
14.1	7.5	16.5	4.7
17.9	4.7	20.8	2.4
25.7	1.0	28.7	0.1

TABLE 4

CLD-2

Adsorption		Desorption	
Moisture content (gm H_2O/100 gm solid)	$-\Delta H_{i(SFL)}$ (cal/gm solid)	Moisture content (gm H_2O/100 gm solid)	$-\Delta H_{i(SFL)}$ (cal/gm solid)
0.0	43.9	6.7	18.2
4.1	28.9	7.7	21.3
4.9	26.7	8.3	17.8
5.6	22.8	11.5	13.5
6.9	21.5	12.8	11.9
7.0	20.3	13.4	10.5
9.4	17.9	16.5	7.5
10.3	16.1	17.0	6.5
14.7	11.6	21.6	4.9
19.3	8.6	31.0	1.6
80.5	0.2	83.3	0.1

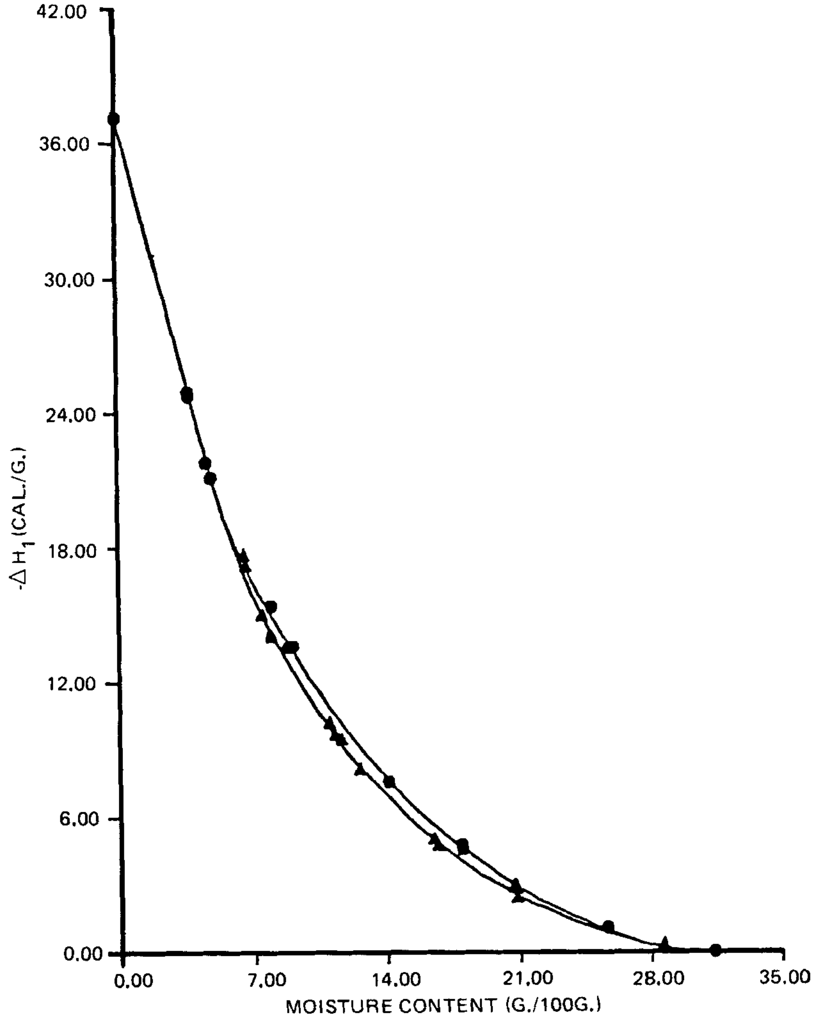

FIGURE 5. Heats of Wetting of Ac-Di-Sol SAmples Containing Various Amounts of Water Vapor for the Adsorption and Desorption Processes.
○ – Adsorption process
△ – Desorption process

One immediately notices in Figures 5 and 6 that the adsorption and desorption curves are not coincidental and, as such, a hysteresis loop is present. This was surprising since Morrison and Dzieciuch[5] did not observe any differences in the

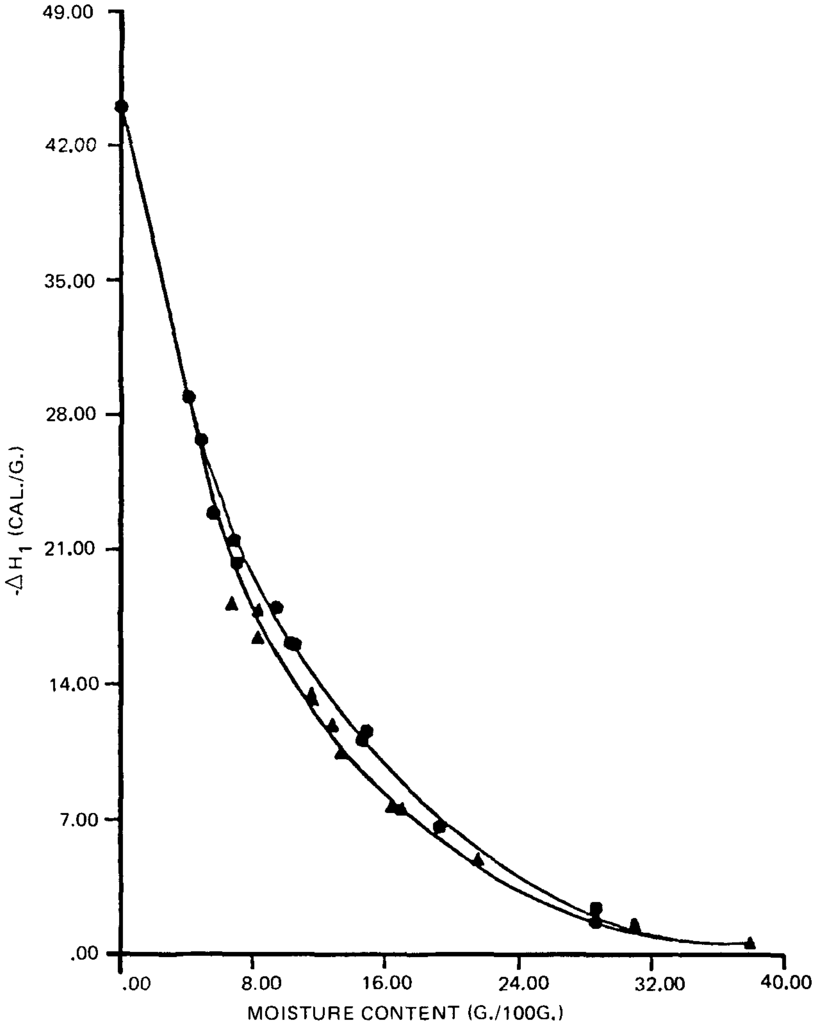

FIGURE 6. Heats of Wetting of CLD-2 Samples Containing Various Amounts of Water Vapor for the Adsorption and Desorption Processes.
○ – Adsorption process
△ – Desorption process

heats of immersion of cellulosic samples which contained equivalent amounts of water vapor irrespective of the process by which the equilibrium water vapor was acquired-adsorption or desorption. The presence of a hysteresis loop can be rationalized by investigating the processes contributing to the total enthalpic change. The change in enthalpy resulting from the immersional process is composed of contributions from two opposing processes. First, a considerable amount of energy is released during interaction between water and the disintegrant's polar functionalities - hydroxyl and carboxyl groups. This exothermic interaction predominates and accounts for the observed negative heats of wetting for the disintegrants investigated. However, if swelling occurs, an endothermic process results due to rupture of solid-solid bonds, and the magnitude of the total negative enthalpy change would be reduced. Thus, the presence of swelling in a sample has been reported to be responsible for the hysteresis loop[3,6].

The heat of adsorption, or the net integral enthalpy, associated with the adsorption process was calculated for both disintegrants using the following equation

$$\Delta H = \Delta H_{I(SL)} - \Delta H_{I} \tag{4}$$

where

$\Delta H_I(SL)$ - is the heat liberated on immersion of the clean, dry solid in water,

ΔH_I - is the heat liberated on immersion in water of the solid containing "n" moles of adsorbed water vapor,

and

ΔH - is the heat liberated due to the adsorption of "n" moles of water vapor.

The entropy change associated with the adsorption process was determined by solving the following equations for entropy:

$$\Delta S = \frac{\Delta H - \Delta G}{T} \tag{5}$$

and

$$\overline{\Delta S} = \frac{\overline{\Delta H} - \overline{\Delta G}}{T} \tag{6}$$

where the bar over the thermodynamic state functions (Equation 6) represents differential values and the thermodynamic functions in Equation 5 are integral values.

Figures 7 and 8 illustrate the relationship between the integral thermodynamic functions and the amount of water vapor adsorbed. As expected, all curves (Figures 7 and 8) show a smooth increase asymptotically approaching a limiting value which corresponds to the maximum moisture adsorbed. However, the representation of these values (ΔG, ΔH and ΔS) in its current form does not differentiate the effect the adsorption of each n mole of water vapor has on the change of the thermodynamic state functions. Therefore, the thermodynamic state functions were differentiated with respect to the amount

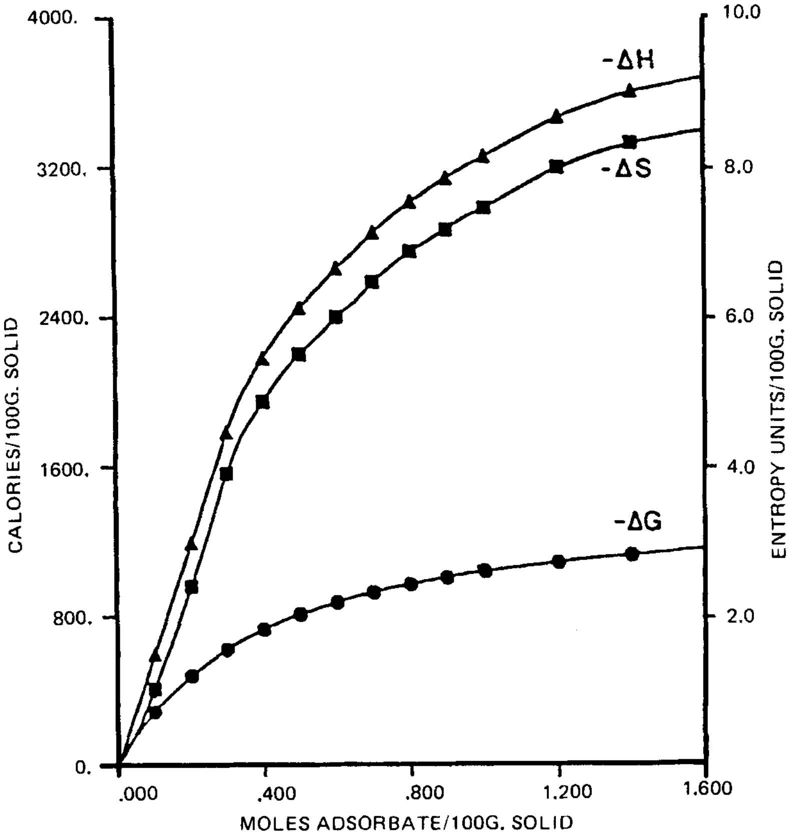

FIGURE 7. Integral Thermodynamic Properties of Adsorption for the Water-Ac-Di-Sol System.

of water vapor adsorbed, and these values are illustrated in Figures 9 and 10.

Because a linear relationship was observed between the heats of immersion, H_i, and the amount of water vapor adsorbed at low moisture contents (Figures 7 and 8), a constant differential enthalpy of adsorption was likewise observed over the same moisture content range. Although there is disagreement over the significance of the constant differential

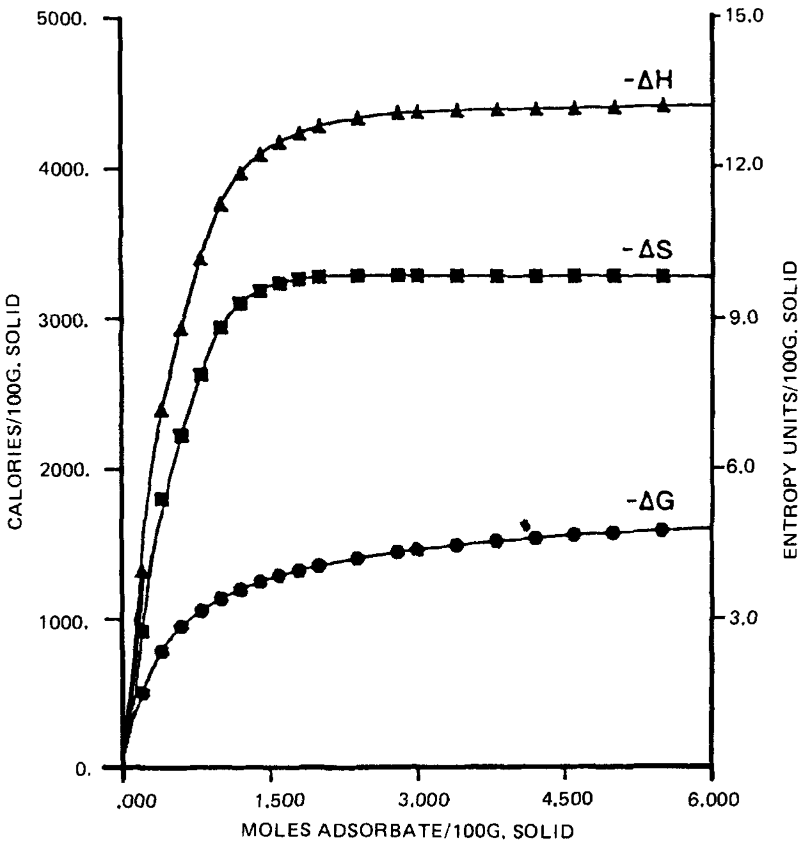

FIGURE 8. Integral Thermodynamic Properties of Adsorption for the Water-CLD-2 System.

enthalpy period[9,10], the energy released on immersion is contingent on the amount of bare surface available for interaction. Therefore, the constant rate period would be expected to extend to completion of the BET monolayer represented in Figures 9 and 10 by a dashed line. If, however, the adsorbed water molecules begin to interact with their adjacent neighbors (latreal adsorbate interactions) and/or the solid swells, the negative differential enthalpies would

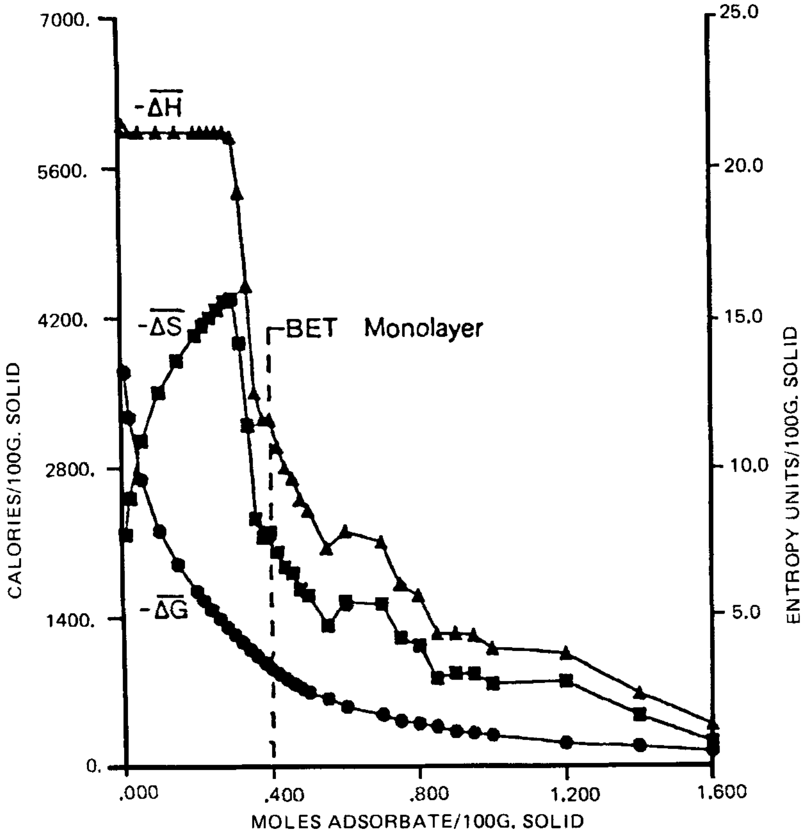

FIGURE 9. Differential Thermodynamic Properties of the Water-Ac-Di-Sol System.

decrease rapidly before completion of monolayer coverage.

Because a zero change in the differential enthalpy indicates that the hydrogen bonds formed between the water and the disintegrants on immersion have an energy equivalent to the hydrogen bond strength of liquid water, the differential enthalpy values in Figures 9 and 10 are in excess, or above, the heat of condensation of liquid water.

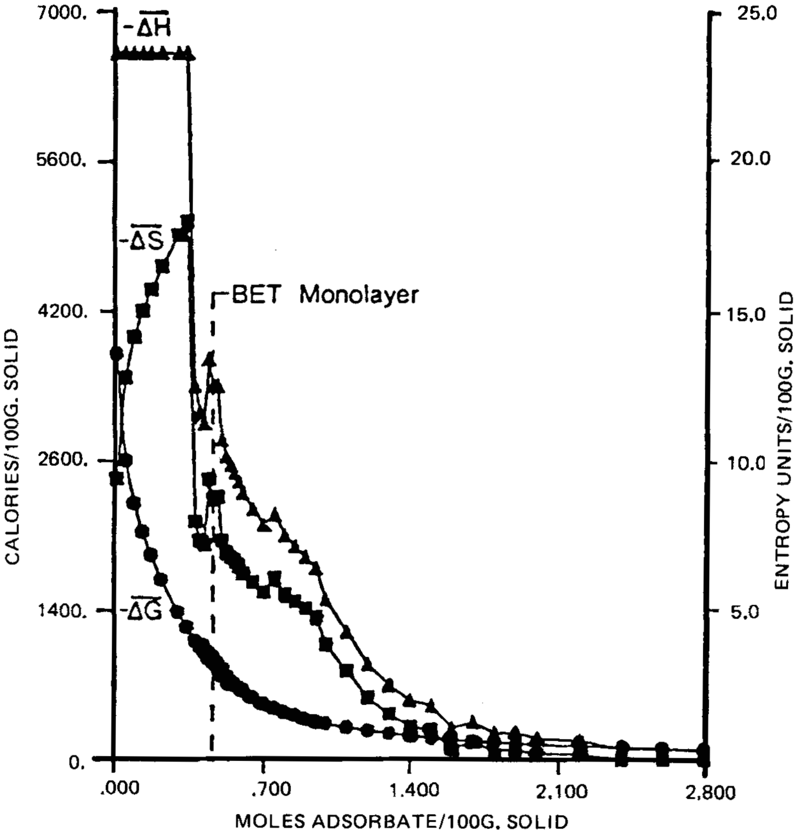

FIGURE 10. Differential Thermodynamic Properties of the Water-CLD-2 System.

An adsorbed water molecule, at low moisture contents (<0.28 and 0.34 moles H_2O/100 grams for Ac-Di-Sol and CLD-2), is likely to form two bonds with the disintegrant's surface. In these ranges, the differential enthalpy has a value of -6.0 kcal/mole and -6.6 kcal/mole as indicated by the constant differential enthalpy levels in Figures 9 and 10. This represents the bond energy in excess of normal hydrogen bonds in water and is due to the formation of two hydrogen bonds between each water molecule and the surface of the solid disintegrant. The excess energies, on a per hydrogen bond basis, are the -3.0 kcal/mole and -3.3 kcal/mole for Ac-Di-Sol and CLD-2, respectively. The total bond energy between water and either Ac-Di-Sol or CLD-2 were calculated by adding the energy for one mole of hydrogen bonds in water (-4.5 kcal/mole) to the experimentally measured excess (-3.0 kcal/mole and -3.3 kcal/mole). These values are -7.5 kcal/mole and -7.8 kcal/mole for Ac-Di-Sol and CLD-2.

Following the constant differential enthalpy, a steep decrease usually occurs followed by an inflection point around the calculated B.E.T. monolayer value. The appearance of the inflection point is due to the transition from a situation where the majority of water molecules interact by forming two bonds with the surface of the disintegrant to a complete monolayer involving one solid-liquid bond per water molecule (entropy effect). The energies of interaction between the adsorbed water vapor and the surface of the disintegrant at the inflection points were -2.1 kcal/mole and -3.15 kcal/mole for Ac-Di-Sol and CLD-2, respectively. Assuming that this entire energy of interaction is in excess of normal hydrogen bond formation in liquid water, the calculated values of interaction between water and either Ac-Di-Sol or CLD-2 were -6.6 kcal/mole and -7.65 kcal/mole.

A comparison of the energies of interaction between water vapor and the surface of the disintegrants at the constant rate period and the corresponding inflection points would be interesting in evaluating the presence of swelling. For example, if swelling occurs, the energy value at the inflection point would be reduced due to consumption of energy to break the solid-solid bonds. This results in an energy of interaction lower than would be obtained if the solid did not swell. Thus, based on the data already reported that Ac-Di-Sol and CLD-2 swell considerably[7], a -0.9 kcal/mole and a -0.15 kcal/mole reduction in interaction energies for Ac-Di-Sol and CLD-2 was not a surprise. Although Ac-Di-Sol consumes 0.75 kcal/mole more energy when compared to CLD-2, this was not unexpected since previously we demonstrated that up to 81.2% relative humidity Ac-Di-Sol exhibited greater swelling tendencies than CLD-2[7].

The differential entropy represents the rate at which the system is moving toward or away from its maximum degree of disorder and is influenced by four factors: 1. the change from three-dimensional, random, molecular motion to two-dimensional, restricted, motion; 2. the ordering of water molecules in the surface film; 3. the separation of water molecules in surface spreading; and 4. the swelling of the solid.

The differential entropy at very low moisture contents is positive and reflects the separation effects predominance on the water molecules. This region is seldom illustrated since data is hard to obtain at extremely low relative humidities. As more moisture is adsorbed, however, the differential entropy will continually decrease and is illustrated in Figures 9 and 10. The continual increasing values of the negative differential entropy is a result of a decrease in the separation effect; in other words, the increase in the negative differential entropy corresponds to a period of accelerated ordering, due primarily to the approach of monolayer completion. Maximum ordering occurs at 0.28 and 0.34 grams of water per 100 grams of Ac-Di-Sol and CLD-2.

Wahba[11] has reported that a maximum ordering will actually occur slightly before monolayer completion because there will undoubtedly be some premature multilayer formation. However, if swelling occurs in the solid, a considerable disturbance of the solid's surface results and dominates the rate of entropy change. This disturbance, or randomization, of the solid's surface would mean that the maximum value of the differential entropy would occur significantly prior to monolayer coverage as can be observed in Figures 9 and 10.

Despite the swelling process and continued adsorption, a small peak is still observed in the two differential entropy curve (Figures 9 and 10) in the region of the B.E.T. monolayer. These peaks attest that the tremendous degree of ordering represented by monolayer completion is a significant event.

The adsorption of water on both cross-linked sodium carboxymethylcellulose disintegrants containing large amounts of moisture is similar in character to the adsorption of water on pure water. Such behavior is characteristic of fibrous solids. Fibers tend to confine the adsorption so that interactions between neighboring layers of adsorbate are inevitable. The heats of wetting of Ac-Di-Sol and CLD-2 containing large amounts of water confirm this belief by showing a negligible change in the negative differential enthalpy.

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