Supplementary Information

Scheme S1 Reaction cascade of the electrophilic attack of the arylnitrenium ions of 2-NBA and 3-NBA on the exocyclic amine in guanine. The numbers correspond to the values in Table S1.

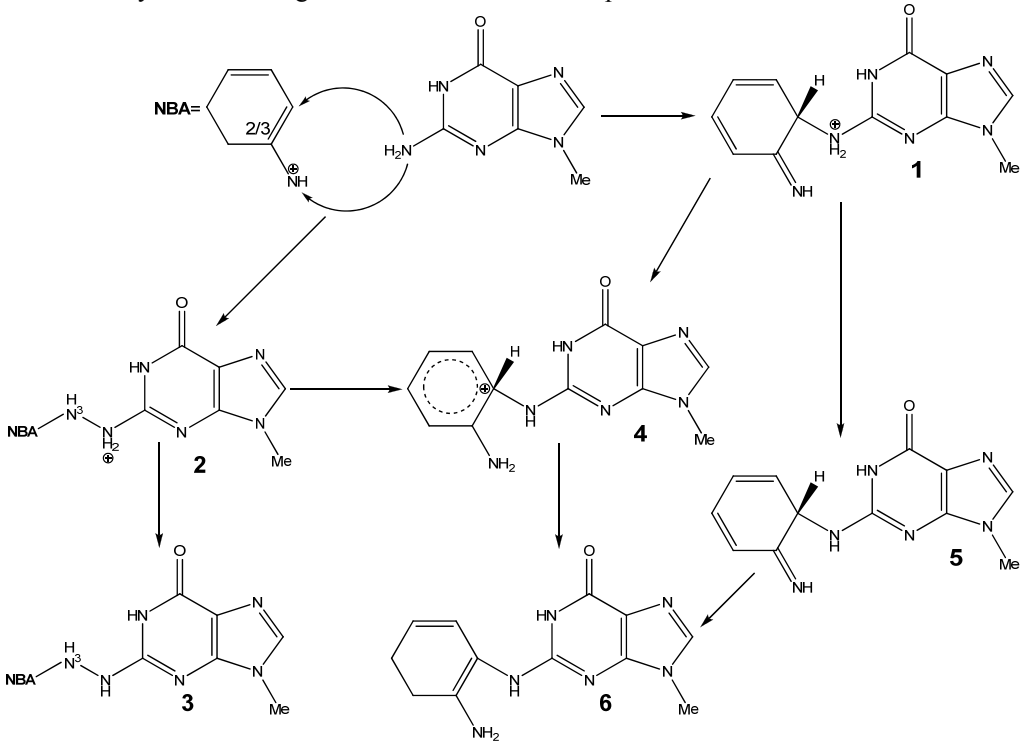

Table S1. The single point and zero point vibrational energies (ZPE) of the structurally optimized molecules in Hartrees (a.u.). The molecular structures are shown in Schemes S1.

System	Energy / a.u.	ZPE / a.u.
Guanine	-582.0514962	0.141935449
Adenine	-506.7892679	0.137285412
3-NBA-ion	-783.7774724	0.218151745
2-NBA-ion	-783.7574330	0.218151745
2-NBA		
1	-1365.842707	0.363921539
2	-1365.816976	0.218151745
3	-1365.491147	0.352240073
4	-1365.874299	-1339.103163
5	-1365.483837	0.352093993
6	-1365.525838	0.352317524
3-NBA		
1	-1365.839419	0.364249973
2	-1365.814527	0.364005853
3	-1365.489293	0.352610664
4	-1365.866542	0.364304875
5	-1365.480049	0.352242034
6	-1365.522357	0.351580264

Scheme S2 Reaction cascade of the electrophilic attack of the arylnitrenium ions of 2-NBA and 3-NBA on C8 in guanine. The numbers correspond to the values in Table S2.

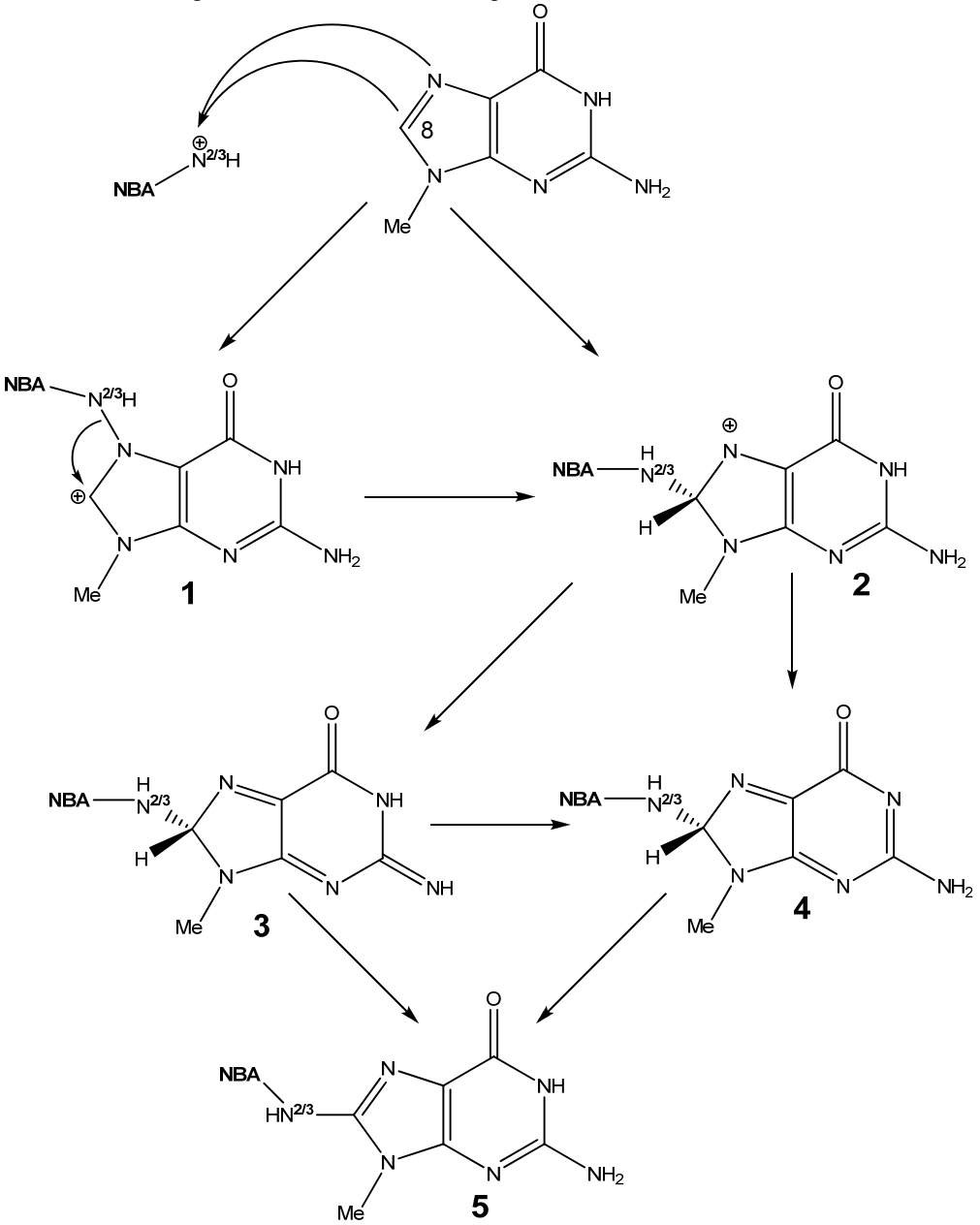

Table S2. The single point and zero point vibrational energies (ZPE) of the structurally optimized molecules in Hartrees (a.u.). The molecular structures are shown in Schemes S2.

System	Energy / a.u.	ZPE / a.u.
2-NBA		
1	-1365.878520	0.364155854
2	-1365.844473	0.363552908
3	-1365.473519	0.351241045
4	-1365.479107	0.35088614
5	-1365.533236	0.351720461
3-NBA		
1	-1365.877760	0.364048991
2	-1365.871653	0.363661733
3	-1365.470659	0.351169476
4	-1365.477525	0.350868493
5	-1365.531843	0.352042032

Scheme S3 Reaction cascade of the electrophilic attack of the arylnitrenium ions of 2-NBA and 3-NBA on C8 in guanine. The numbers correspond to the values in Table S3.

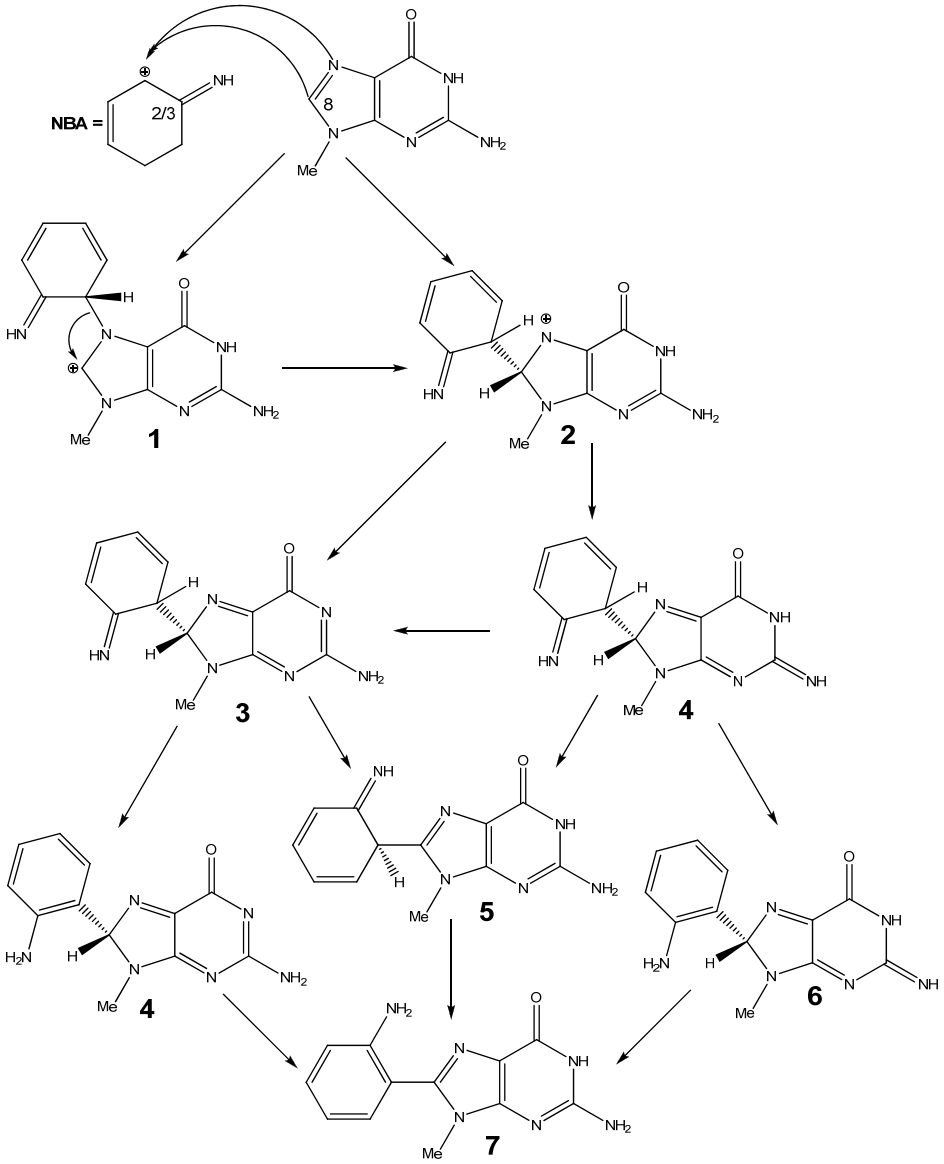

Table S3. The single point and zero point vibrational energies (ZPE) of the structurally optimized molecules in Hartrees (a.u.). The molecular structures are shown in Schemes S3.

System	Energy / a.u.	ZPE / a.u.
2-NBA		
1	-1365.888817	-1339.117396
2	-1365.854094	-1339.083354
3	-1365.454025	0.350534177
4	-1365.447653	0.350864572
5	-1365.482760	0.35125281
6	-1365.499636	0.351933208
7	-1365.475438	0.351542028
8	-1365.539543	0.352447918
3-NBA		
1	-1365.887879	0.363861734
2	-1365.850059	0.363580360
3	-1365.450543	0.350731237
4	-1365.442922	0.350967514
5	-1365.488205	0.350991043
6	-1365.495455	0.352062620
7	-1365.477958	0.350854768
8	-1365.537356	0.352333211

Scheme S4 Reaction cascade of the electrophilic attack of the arylnitrenium ions of 3-NBA on the exocyclic amine in adenine. The numbers correspond to the values in Table S4.

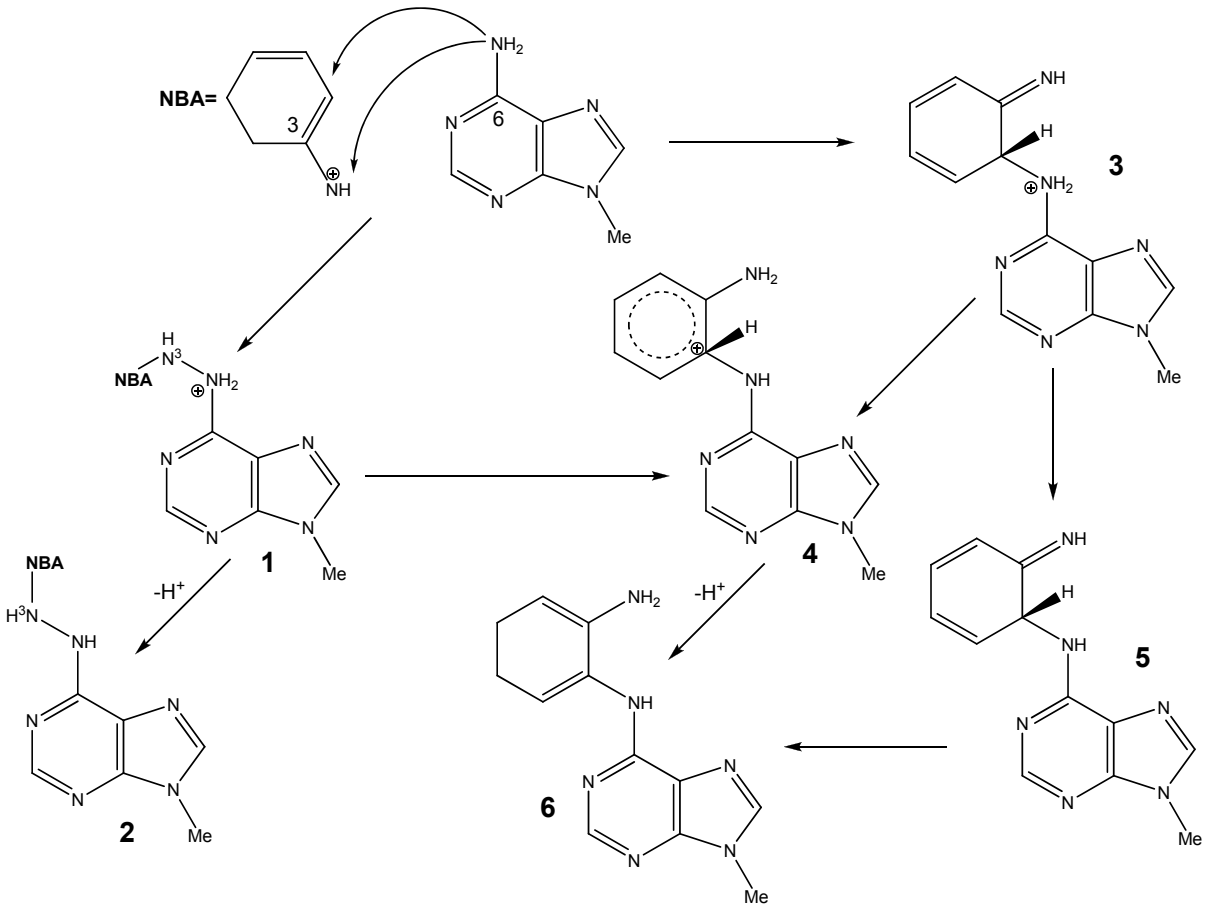

Table S4. The single point and zero point vibrational energies (ZPE) of the structurally optimized molecules in Hartrees (a.u.). The molecular structures are shown in Schemes S4.

System	Energy / a.u.	ZPE / a.u.
1	-1290.573007	0.360959750
2	-1290.218199	0.347970431
3	-1290.588693	0.359767584
4	-1290.617806	0.360349942
5	-1290.219449	0.347463564
6	-1290.262826	0.347933176

Scheme S5 Reaction cascade of the electrophilic attack of the arylnitrenium ion of 3-NBA on C8 in adenine. The numbers correspond to the values in Table S5.

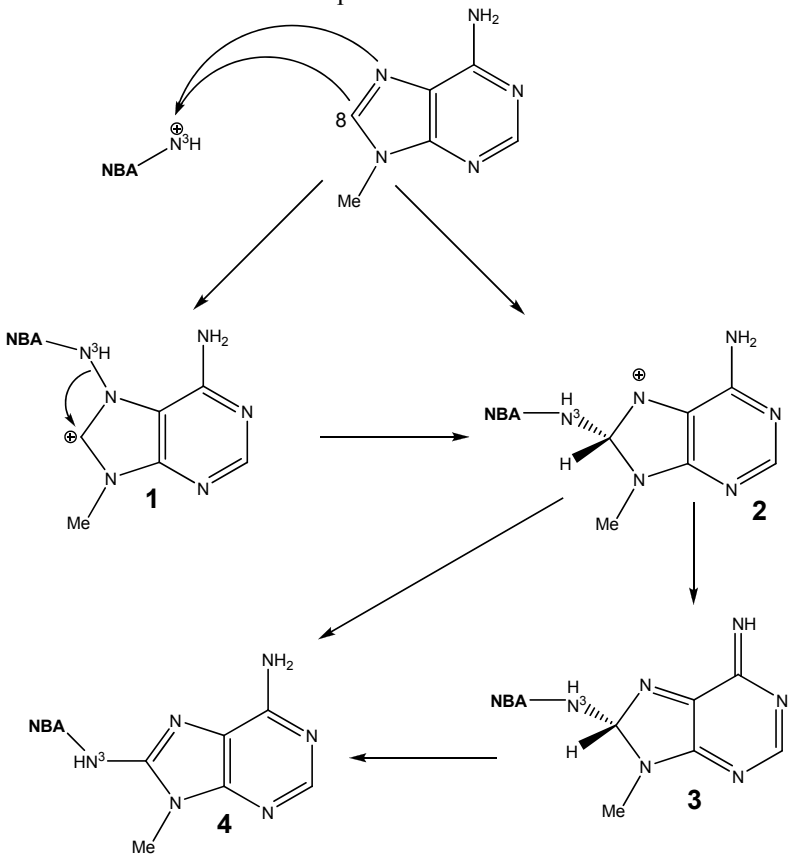

Table S5. The single point and zero point vibrational energies (ZPE) of the structurally optimized molecules in Hartrees (a.u.). The molecular structures are shown in Schemes S5.

System	Energy / a.u.	ZPE / a.u.
1	-1290.600206	-1265.304442
2	-1290.558000	0.358983264
3	-1290.189473	0.346716499
4	-1290.187517	0.346128259

Scheme S6 Reaction cascade of the electrophilic attack of the arylnitrenium ion of 3-NBA on C8 in adenine. The numbers correspond to the values in Table S6.

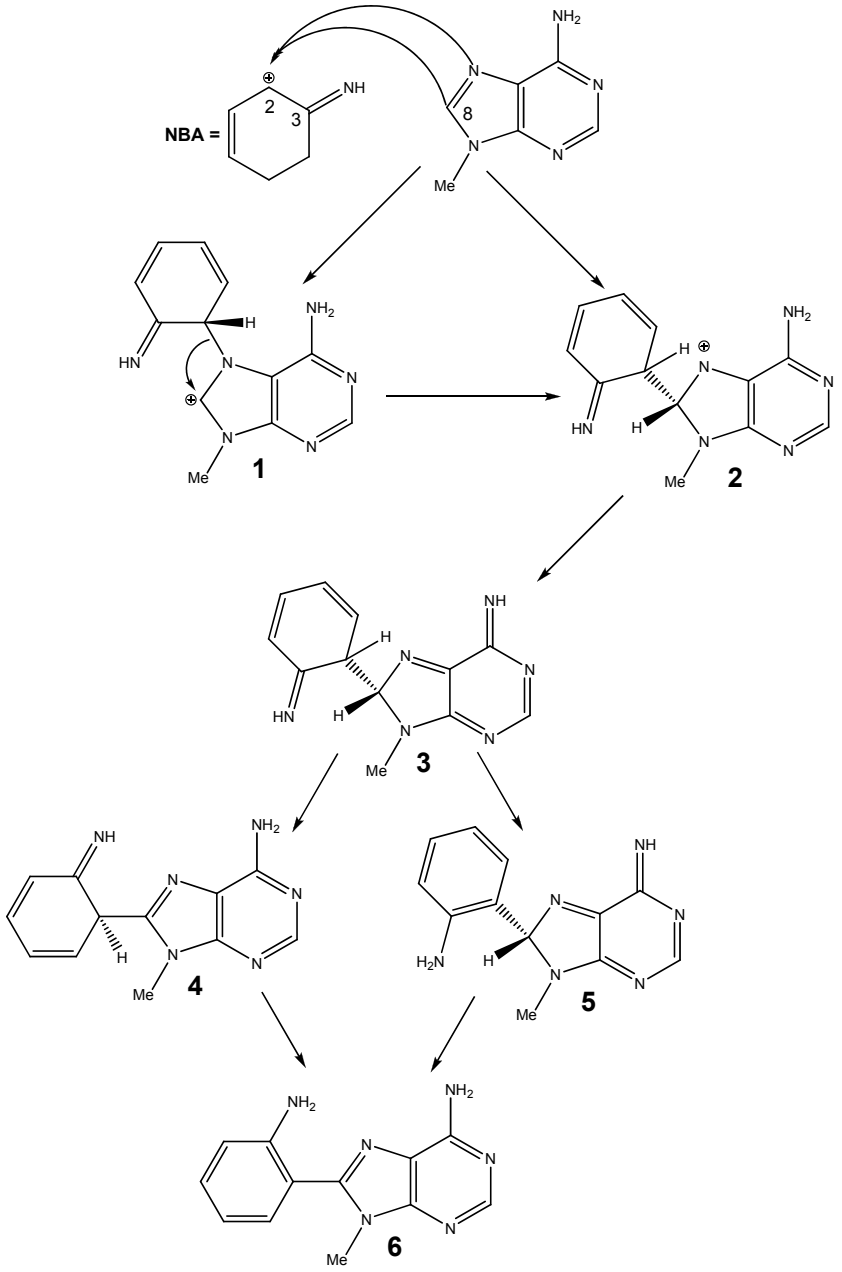

Table S6. The single point and zero point vibrational energies (ZPE) of the structurally optimized molecules in Hartrees (a.u.). The molecular structures are shown in Schemes S6.

System	Energy / a.u.	ZPE / a.u.
1	-1290.604128	0.359641112
2	-1290.565639	0.358703850
3	-1290.162284	0.346432183
4	-1290.232652	0.347085130
5	-1290.196046	0.346065514
6	-1290.274657	0.347684154