David Peters

Food Chemistry: An Organic Perspective

Baran Group Meeting
10/5/19

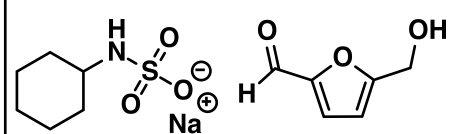
Food Chemistry:
The study of the chemical processes, both biological and abiotic, that occur in food substances during processing, handling, and consumption.

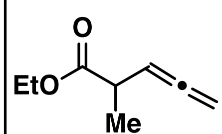
Food Chemists:
solve problems of the above processes with the use of change of food components, their ratios, and conditions

Goup Meeting DOES NOT Include:
- in-depth coverage of “food chemistry”
- concepts of food processing
- discussion of nutrition
- any official health advice regarding diet

Goup Meeting Includes:
- organic chemistry-related topics regarding food
- major reactions during cooking
- flavor addititives
- food coloring additives
- vitamins and minerals (brief)
- other common additives & preservatives
- fun & interactive chemical exploration

WHY HAVE YOU EATEN THESE?!





Major references for this group meeting:

- Nursten, H. The Maillard Reaction **2005**. Cambridge, UK. The Royal Society of Chemistry.
- Baht, S.V., *et. al.* Chemistry of Natural Products **2005**. New York, NY, Springer.
- Ullman’s Encyclopedia of Industrial Chemistry **2002**. Wiley-VCH.
- Porter, J. Cooking for Geeks **2010**, Sebastapol, CA. O’Rielly Media.
- www.FDA.gov
- www.thegoofscentscompany.com
- www.femaflavor.org

TODAY’S CASE STUDIES

- *What was in Pepsi Blue?*
- *What exacty does Phil drink after the gym?*
- *What is in those jelly beans?*

Pepsi Blue®

Muscle Milk®

Nutrition Facts
Serving Size 11 fl. oz. (330 mL)
Servings Per Container 1

Amount Per Serving	
Calories 100	Calories from Fat 10
	% Daily Value*
Total Fat 1g	**2%**
Saturated Fat 0.5g	**3%**
Trans Fat 0g	
Polyunsaturated Fat 0g	
Monounsaturated Fat 0g	
Cholesterol 10mg	**3%**
Sodium 230mg	**10%**
Potassium 640mg	**18%**
Total Carbohydrate 5g	**2%**
Dietary Fiber 3g	**12%**
Sugars 0g	
Protein 20g	**40%**

Vitamin A 20%	• Vitamin C 20%
Calcium 30%	• Iron 20%
Vitamin D 20%	• Vitamin E 20%
Thiamin 20%	• Riboflavin 20%
Niacin 20%	• Vitamin B6 20%
Folate 20%	• Vitamin B12 20%
Biotin 20%	• Pantothenic Acid 20%
Phosphorus 40%	• Iodine 20%
Magnesium 20%	• Zinc 20%
Copper 20%	• Chromium 50%

*Percent Daily Values are based on a 2,000 calorie diet. Your daily values may be higher or lower depending on your calorie needs:

	Calories	2,000	2,500
Total Fat	Less than	65g	80g
Saturated Fat	Less than	20g	25g
Cholesterol	Less than	300mg	300mg
Sodium	Less than	2,400mg	2,400mg
Potassium		3,500mg	3,500mg
Total Carbohydrate		300g	375g
Dietary Fiber		25g	30g
Protein		50g	65g

Calories per gram:
Fat 9 • Carbohydrate 4 • Protein 4

INGREDIENTS: WATER, MILK PROTEIN ISOLATE, CALCIUM CASEINATE (MILK), SODIUM CASEINATE (MILK), LESS THAN 1% OF: ALKALIZED COCOA POWDER, SOLUBLE VEGETABLE FIBER, NATURAL AND ARTIFICIAL FLAVORS, MALTODEXTRIN, CELLULOSE GUM AND GEL, DIPOTASSIUM PHOSPHATE, MAGNESIUM PHOSPHATE, POTASSIUM CHLORIDE, SODIUM HEXAMETAPHOSPHATE, SODIUM PHOSPHATE, CARRAGEENAN, ACESULFAME POTASSIUM, CANOLA OIL, SUNFLOWER OIL, POTASSIUM CITRATE, ASCORBIC ACID, CALCIUM PHOSPHATE, FERRIC PYROPHOSPHATE, SUCRALOSE, DL-ALPHA TOCOPHERYL ACETATE, D-CALCIUM PANTOTHENATE, NIACINAMIDE, ZINC OXIDE, COPPER GLUCONATE, VITAMIN A PALMITATE, PYRIDOXINE HYDROCHLORIDE, THIAMINE MONONITRATE, RIBOFLAVIN, CHROMIUM CHLORIDE, FOLIC ACID, BIOTIN, POTASSIUM IODIDE, CHOLECALCIFEROL, CYANOCOBALAMIN.

PRODUCED FOR CYTOSPORT, INC. WALNUT CREEK, CA 94597 ©CYTOSPORT, INC.

MUSCLE MILK® LIGHT SHAKES 1G FAT, 100 CALORIES PER SERVING; GENUINE MUSCLE MILK® SHAKES 4G FAT, 130 CALORIES PER SERVING.

GLUTEN FREE Ⓤ D

Jelly Belly®

50 Flavour Assortment Jelly Beans

Nutrition Information	Typical value per 100 g
Energy	1530 kJ / 360 kcal
Fat	0 g
of which	
Saturates	0 g
Carbohydrates	90 g
of which	
Sugars	60 g
Protein	0 g
Salt	0.06 g

INGREDIENTS: Sugar, glucose syrup, modified cornstarch, acidity regulators (E270, E296, E325, E330, E331, E334), flavourings, strawberry puree, colours (E100, E150a, E153, E160a, E162, E171), fruit and vegetable concentrates (carrot, apple, spirulina, black currant, pumpkin, purple sweet potato, hibiscus, radish, grape), raspberry puree, glazing agents (E901, E903, E904), blueberry puree, banana puree, blackberry puree, peach puree concentrate, chocolate (sugar, cocoa mass, cocoa butter), lemon puree, passionfruit juice concentrate, orange puree, grape juice concentrate, mango puree, apple juice concentrate, cocoa powder, lychee juice concentrate, pear juice concentrate, kiwi juice concentrate, tangerine juice concentrate, pomegranate juice concentrate, mango juice concentrate, watermelon juice concentrate, dried coconut powder, grapefruit juice concentrate, cherry juice concentrate, pineapple juice concentrate, salt, cantaloupe powder, lime juice concentrate, freeze-dried soluble coffee, tapioca dextrin, vanilla beans, ground cinnamon.

Foods largely in 3 categories:
- carbohydrates
- proteins
- lipids

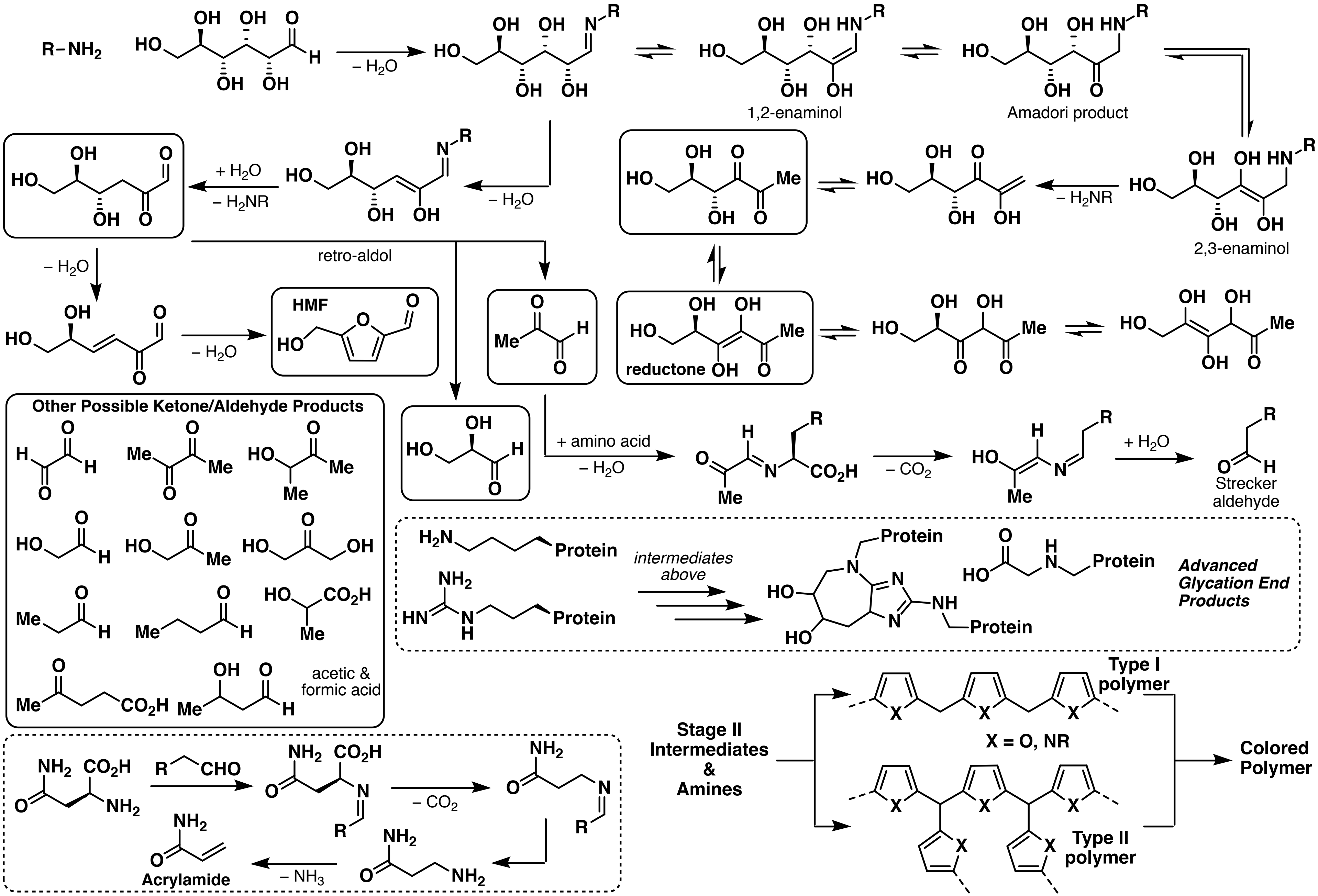
Should there be more?!
- an ever-expanding variety of new chemotypes in food

Food and Drug Administration (FDA):
- sets guidelines, allowances and tollerances for food products
- nothing is ever permanently approved
- “Natural”= original source is natural
- “Artificial” = source is not food-based
- GRAS = generally recognized as safe

EU Food Additive #’s:

E100 to E199: color additives
E200 to E299: preservatives
E300 to E399: antioxidants & pH reg.
E400 to E499: thickeners, stablizers & emulsifiers
E500 to E599: anti-caking
E600 to E699: flavor enhancers
E700 to E799: antibiotics
E900 to E999: miscellaneous
E1000 to E1599: additional chem.

The Maillard Reaction



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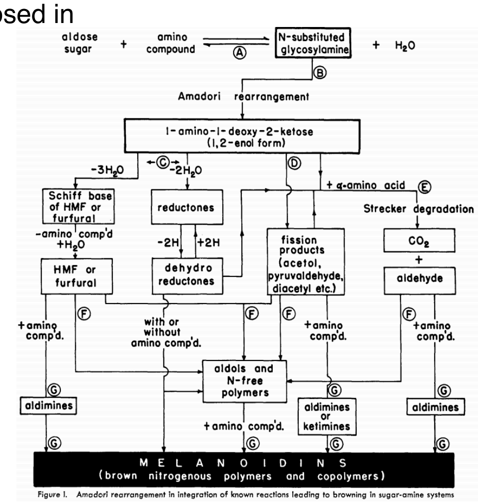
The Maillard Reaction

- term used for non-enzymatic browning of food products when amines and sugars are involved
- First described by Louis-Camille Maillard in 1912 while he is trying to make peptides w/ amino acids and reductants
- *The "reaction" is a mind boggling complex mixture of hundreds of reactants, reactions, and products – hard to study*
- schematic pathway of reactions proposed in 1953 by John E. Hodge
- divided the process into three stages

I. Initial Stage (colorless)
  A. amine-sugar condensation
  B. Amadori rearrangement

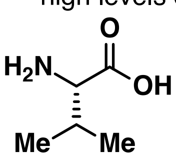
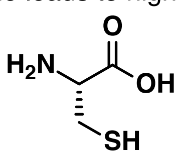
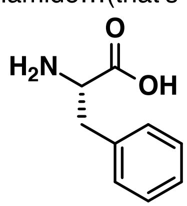
II. Intermediate Stage (light yellow)
  C. sugar dehydration
  D. sugar fragmentation
  E. amino acid degradation

III. Final Stage (highly colored)
  F. aldol condensation
  G. adehyde-amine polymerization/ heterocycle formation



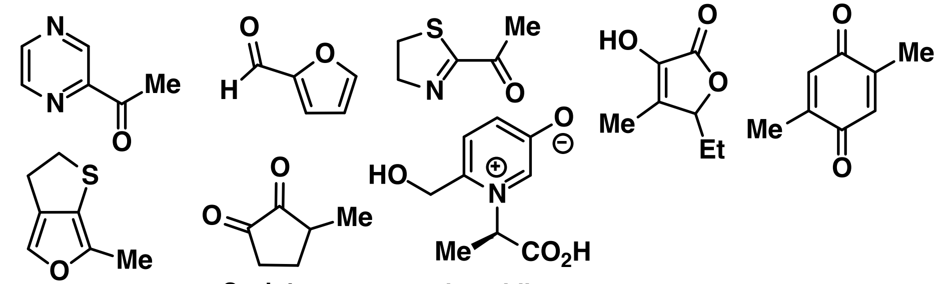
Hodge, *J. Agric. Food Chem.* **1953**, *1*, 928.

Starting materials:
- N-terminal amines, lysine side chain, or any other amine!
- taste and smell mostly independent of sugars present
- taste and smell dependent upon amino acids present - ***Strecker aldehydes***
- high levels of asparagine leads to high levels of acrylamide…(that's bad)

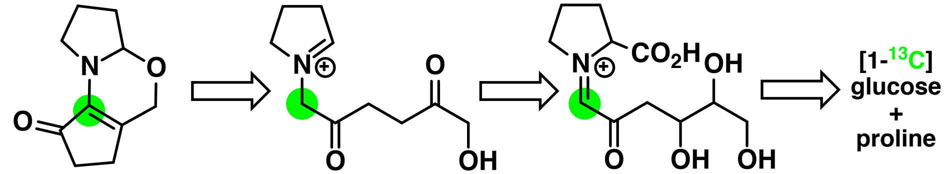
 = bread  = meat/ crackers  = floral

Progress:
- becomes noticable ~154 ºC (310 ºF); cooking below this = no browning
- extent of reaction is depended upon time, temperature, pH and pressure
- pH can change the dominant reaction pathway
- 1,2-enol @ low pH; 2,3-enol @ high pH - effects product distribution
- progress inhibited by bisulfite (used in food industry)

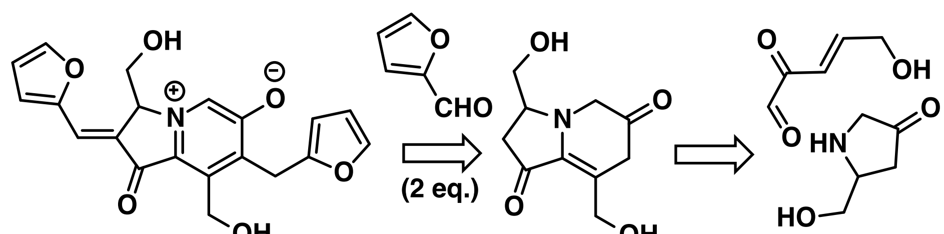
Products:
- produces heterogeneous polymers, volatile hydrocarbons, complex heterocycles
- the colored polymeric structures are called **melanoidins**
- molar mass of polymers increases w/ *temperature* NOT *time*
- those above 3 kDa are generally insoluble
- melanoidins are 25% of the dry mass of coffee
- numerous other products can be benificial or detrimental to taste
- carcinogenic/mutagenic compounds are genereally product of very high temperatures and long reaction times
- acrylamide is result of high temp. and high concentration of asparagine



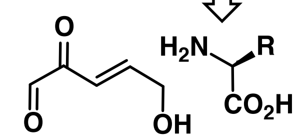
kahweofuran **Cyclotene** **alapyridine**



maltoxazine



indolizinium-6-olate derivative



- **kahweofuran** is in coffee; gives a roasted/smoky aroma
- **cyclotene** is an important caramel odorant
- **alapyridine** increases sensitivity to sweetness ~16x
- **maltoxazine** is in dark malt (stouts/porters); 10 mg/kg
- **indoliziniums** are extremely bitter and are undesirable

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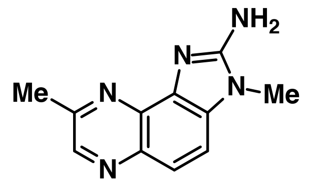
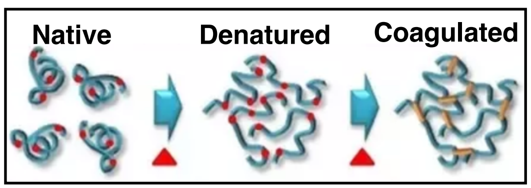
Food Item	acrylamide (ng/g)
Potatoes (raw)	n.d.
Potatoes (boiled)	n.d.
Potato chips (fried)	3500
Potato chips (fried; overcooked)	13000
Rye crackers	4000
Breakfast cereal	1400
Breakfast cereal (rice-based)	250



IQx
[highly mutagenic]

Proteins in Food:

- where we get the majority of our amino acids to turn into our own proteins
- denatured proteins are a staple of human food
- heat and pH and mechanical aggitation are major the factors in destroying native proteins before consumption
- denatured/coagulated proteins change in texture, solubility, color, solublizing properties, etc. (nearly all physio-chemical properties)
- the change in properties is often utilized to create food properties
    - *eg.*– properly denatured egg whites are emulsifiers of air
- temperatures of cooking are determined by this process
    - *eg.*– people prefer meat *with* actin but *without* myosin

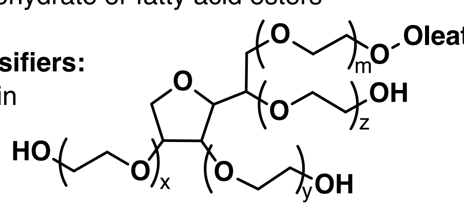


Temperatures of Denaturation:
Egg whites: 61 °C (122 °F)
Myosin: 50 °C (122 °F)
Actin: 65.5 °C (150 °C)

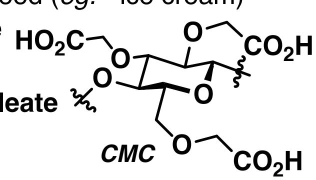
Emulsifiers/Emulsification:

- emulsifiers act as the border between oil and water
- help ***create and maintain*** the texture and from of food (*eg.* - ice cream)
- Must be a ambiphillic molecule to affect its purpose
- generally carbohydrate or fatty acid esters

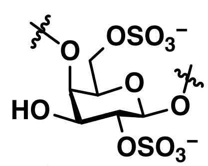
Common Emulsifiers:
- soy/egg lecitin
- canola oil
- carrageenan
- polysorbates
- carboxymethylcellulose (CMC)
- guar gum (galactose/manose; branched)
- xanthan gum (glucose/manose; branched)



polysorbate-80



CMC

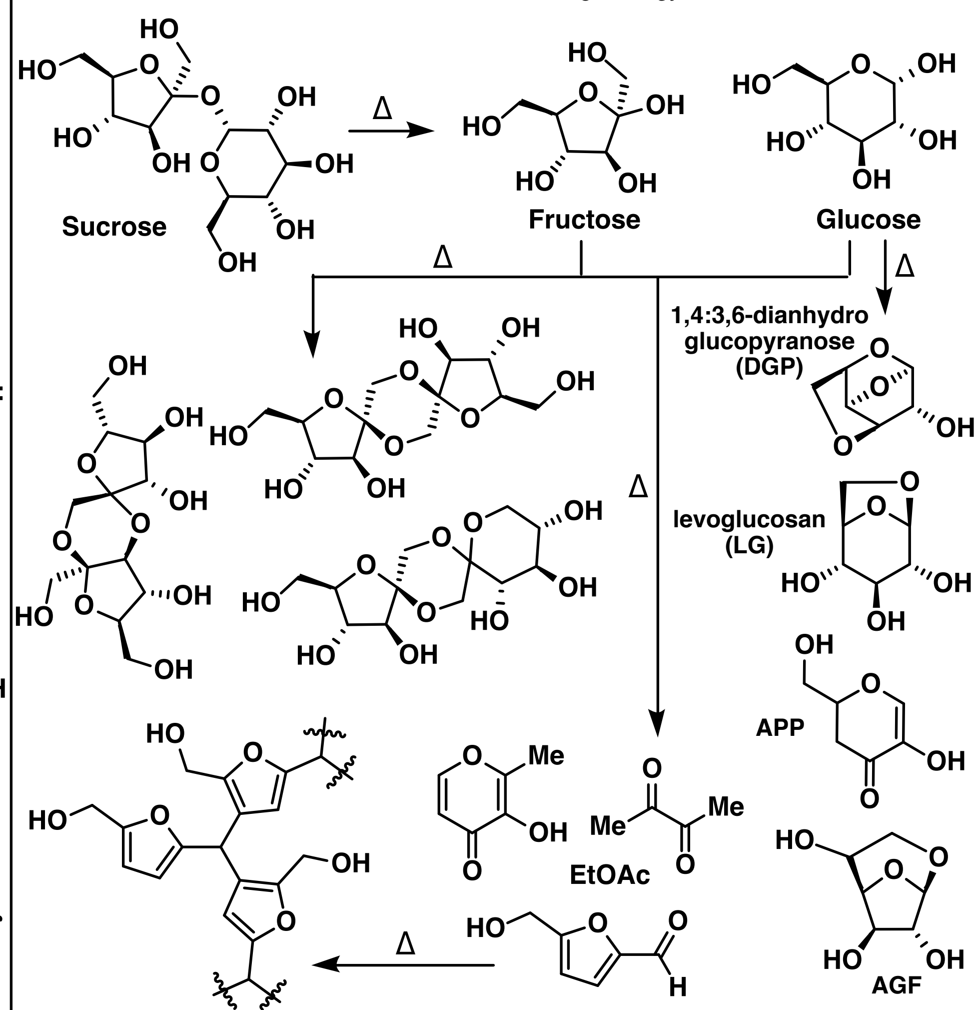


Carrageenan

Caramelization

- term used for the non-enzymatic browning of sugars (alone)
- the process is a form of pyrolysis (irreversible thermal decomposition w/o combustion)
- products/reaction intermediates are very similar to those of Maillard Rection
- Begins ~150 °C; Most noticable between 160-200 °C (320-400 °F)
- low temp. = light colors
- high temp. = dark colors
- medium temp. = rich flavors
- most studied model is that of sucrose

Yang, *Energy and Fuels* **2017**, *31*, 8291.



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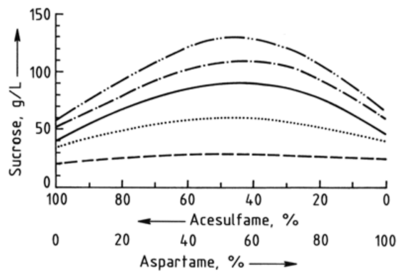
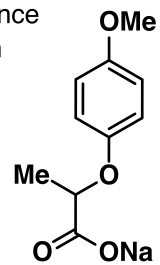
Artificial Sweeteners:

- saccharin was the first (1884) - WWI and WWII helped its poularity
- natural non-caloric sweeteners used throughout world previously
- originaly used mainly by diabetics and was taxed to protect sugar industry
- obesity & health affects of high-sugar diets has changed perspective/demand
- sweetness is measured as a relative to dilute sucrose (~0.1g/L)
- all sweeteners have an asymtotic sweetness w/ concentration
- many have "off tastes" - circumvented via blends of sweeteners
- blended sweeteners are generally synergistic in sweetness
- may actually combat tooth decay
- most sweeteners are found by chance
- very little sweetness-SAR is known

$Pb(OAc)_2$
"Sugar of Lead"



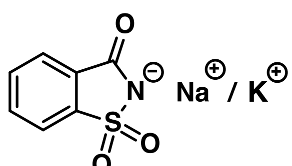


Lactisole
[anti-sweetener]

Sweetness Intensity	
sucrose	1
sodium cyclamate	35
stevioside	160
acesulfame K	200
aspartame	200
rebaudioside A	250
neohesper. DHC	330
sodium saccharin	2500
neotame	8000
advantame	47800

Saccharin:

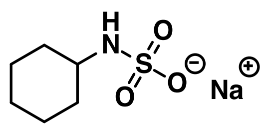
- 1.0 kg/L solubility in H_2O (Na-salt) – acidic in soln.
- metallic/bitter aftertaste
- fully excreted unmetabolized
- used in concert w/ other sweeteners and sugar
- used in weaning to feed and nickel elctroplating
- ADI: 5 mg/kg/day



Saccharin

Cyclamate:

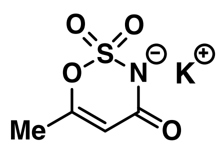
- accidentally discovered in 1937
- Na salt decomposes at 260 ºC
- generally used as 10:1 (cyclamate:saccharin)
- banned in US due to erroneous reports of toxicity
- used in most countries (Canadian Sweet N'Low)



Cyclamate

Acesulfame K

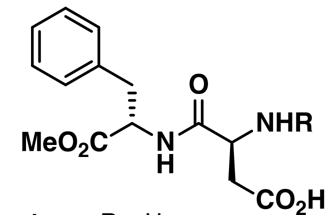
- high temp stability; decomp. at 225 ºC
- metalic taste at high concentrations
- used in concert with numrous others
- extremely soluble in water
- highly absorbed and rapidly excreted unchanged
- ADI: 16 mg/kg/day



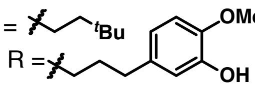
Acesulfame K

Aspartame (and other "tames")

- delayed & lasting sweetness; slight bitterness
- ~4 kcal/g
- aspartame accidentaly discovered in 1965
  - solubility is highest at pH = 2.2
  - maximum stability at pH = 4.0
  - releases methanol and forms DKP
  - ADI: 50 mg/kg/day
  - sweetness decreases after certain [conc.]
- other "tames" do not form DKP
- last two developed based on SAR

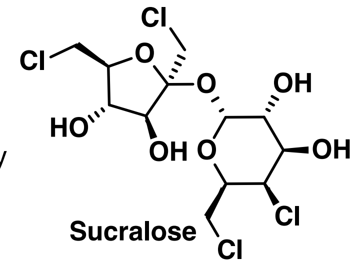


Aspartame: R = H
Neotame: R =
Advantame: R =



Sucralose:

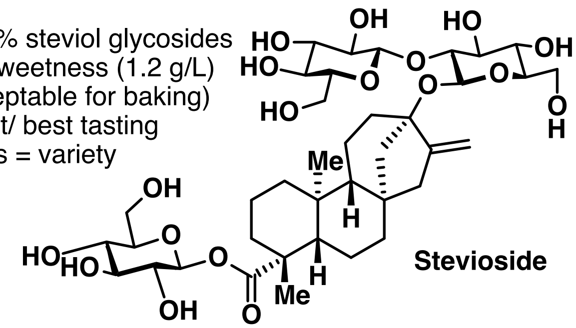
- 1976 discovery after discovery that halogenation increases carbohydrate sweetness
- melts at 125 ºC w/ decomposition
- 283 g/L solubility in water
- slight delay (blends help); lasting effect; generally good
- made from sucrose through PG-heavy synthesis



Sucralose

Steviol glycosides (stevia):

- GMO leaves contain up to 20% steviol glycosides
- just soluble enough for high sweetness (1.2 g/L)
- stable at elevated temp. (acceptable for baking)
- rebaudioside A is the sweetest/ best tasting
- variety of extraction conditions = variety of product qualities
- licorice-like aftertaste
- No ADI; has FDA GRAS



Stevioside

Food Flavorings:

- food flavorings can be both natural and artificial in nature
- flavor chemistry is the field dedicated to crafting flavors wtih the available ingredients and analytical tools
- flavor chemists train in apprenticeships for 2-5 years; after bachelors
- to create a desired artificial flavor there are generally upwards of 10 different additives
- many artificial flavor additives are sold/used as the racemate
- single compounds are usually present in ~5.0 to 0.1 ppm
- many compounds used as both scents (olfactory) and tastes (gustatory)

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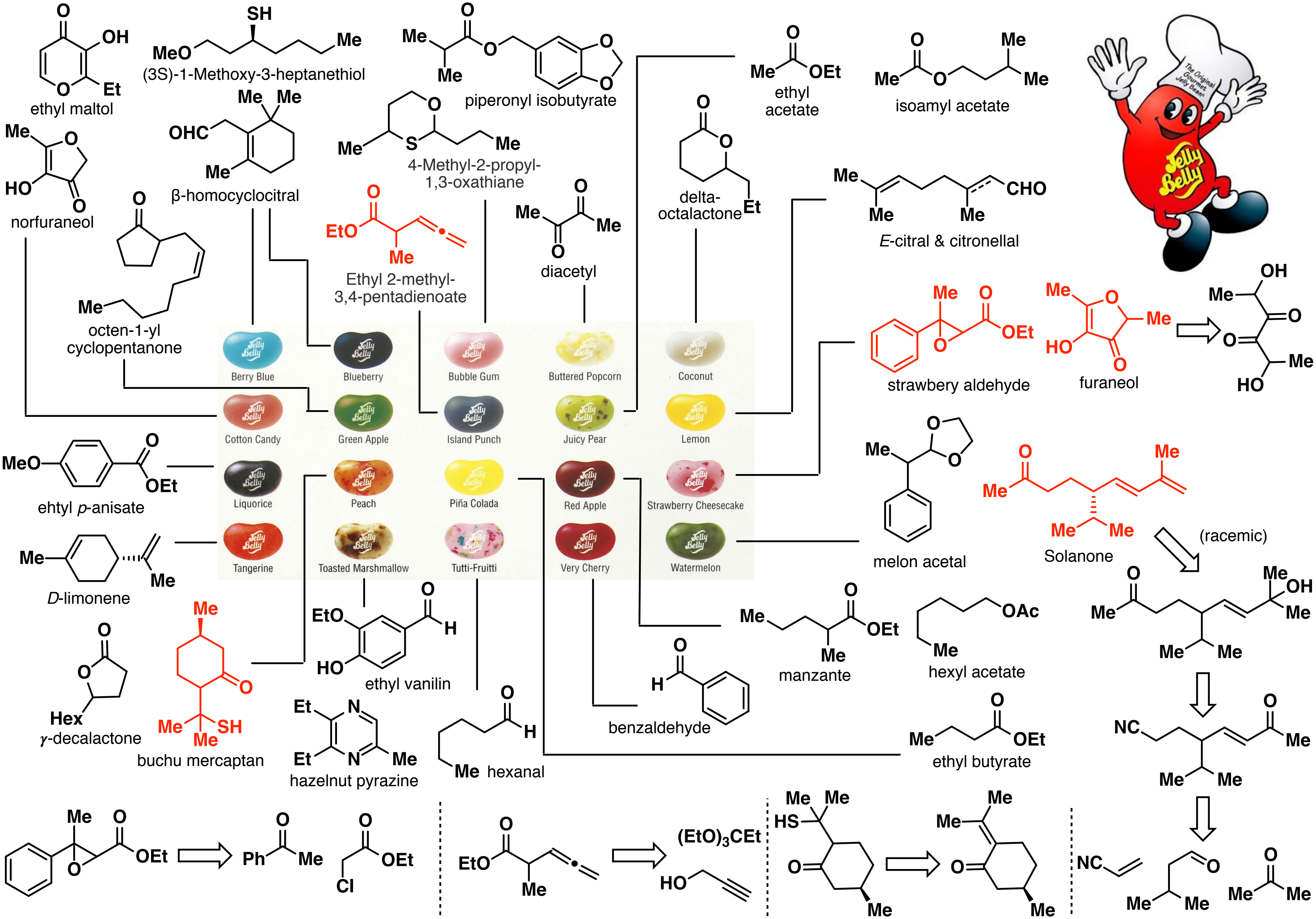
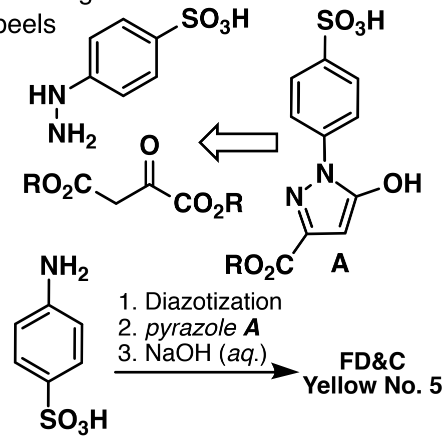


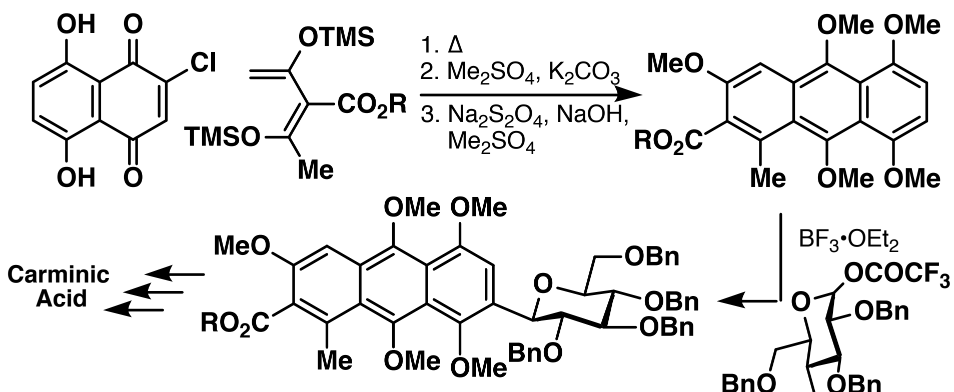
Food Color Additives

- FDA has two categories of colorants: ***Certified*** and ***Exempt***
- **certified *dyes*** are artificial; must be batch-certified
- 16.5 million pounds were certified 2002
- ***exempt dyes*** are derived from natural sources (no certification required)
- ***Lakes*** are dyes mixed with aluminum/calcium and precipitated
- lakes color by dispersion; usually used in oily foods/coatings
- dozens of additional color additives approved for drug, cosmetic, and medical device use (both natural and artificial)
- artificial food dyes originally from the dyestuff industry
- food dyes make up a small fraction of the dyestuff industry
- numerous suggestions that artificial dyes cause hyperactivity in children, cancer, mutagenicity – largely unproven for current set
- Orange B only for use in hotdog/sausage casings
- Citrus Red No. 2 only for use in orange peels

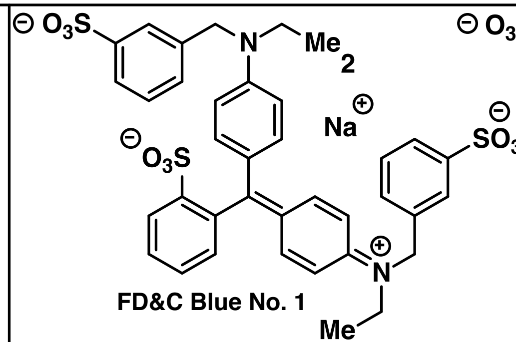
FDA "Exempt" Food Color Additives

annatto extract	Fe^{II} gluconate/lactate
beet powder	Fruit/vegetable juice
canthaxanthin	riboflavin
caramel	carmine
beta-carotenes	cochineal extract
carrot oil	paprika/paprika extract
TiO_2	saffron
mica	spirinula extract
Na Cu chlorophyllin	tomato lycopene
toasted cottonseed	tumeric
iron oxide	tumeric oleoresin

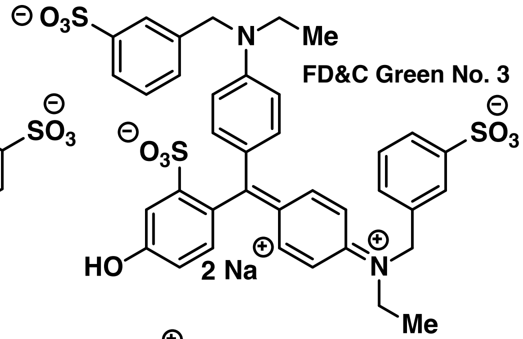




J. Chem. Soc., Perkins Trans. 1 **1999**, 575.



FD&C Blue No. 1



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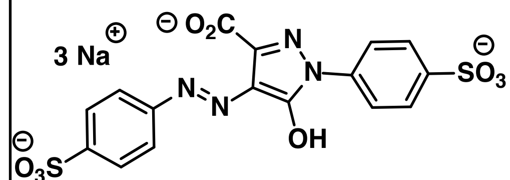
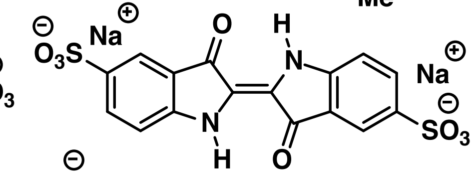
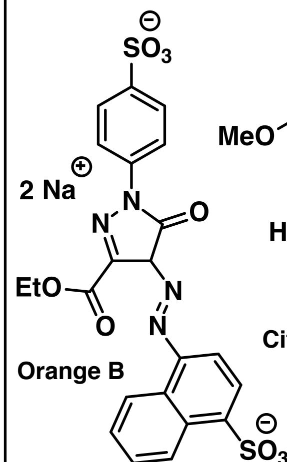


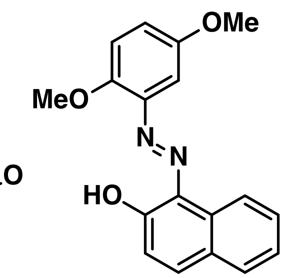
FD&C Yellow No. 5



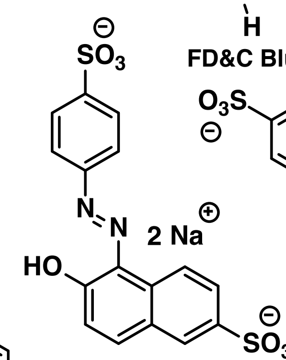
FD&C Blue No. 2



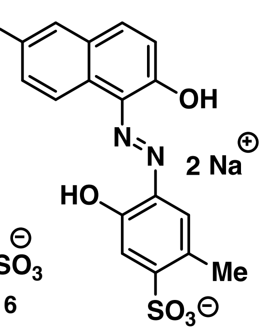
Orange B



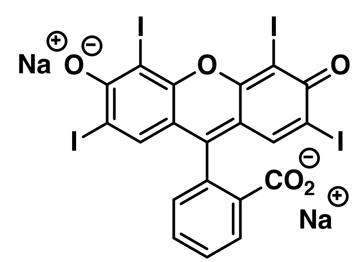
Citrus Red No. 2



FD&C Yellow No. 6

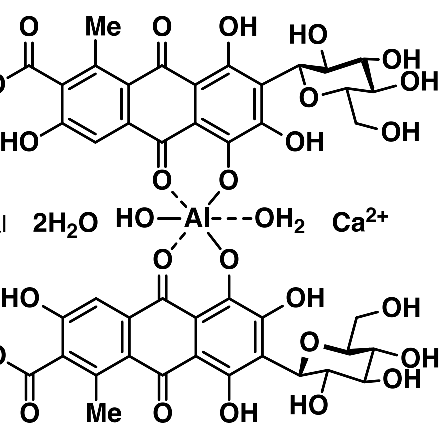


FD&C Red No. 40



FD&C Red No. 3

Carmine (cochineal extract)



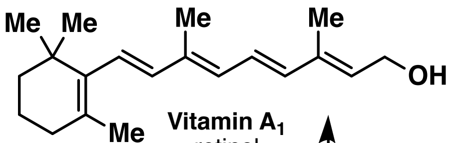
David Peters # **Food Chemistry:** An Organic Perspective **Baran Group Meeting** **10/5/19**

Vitamins

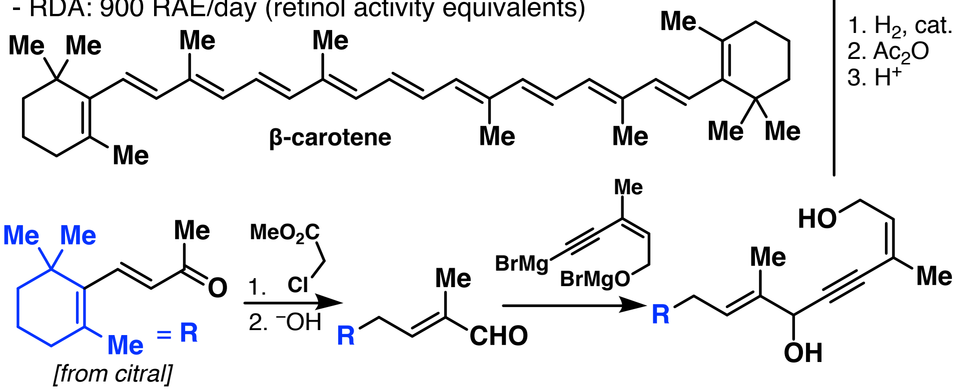
- organic molecules that are essential micronutrients that humans (or other organisms) need for proper metabolic function
- diseases attributed to deficiency in each one
- often derived from natural sources in one's diet
- supplementation and fortification provide full amounts where gaps may occur

Vitamin A (retinoids):

- oxygen and light (*E/Z*) sensitive
- most are interconvertable via enzymes
- 90% stored in liver (as palmitate)
- generally cosidered as antioxidants
- important for epithelial growth
- sensitivity in isolation means current access is synthetic
- RDA: 900 RAE/day (retinol activity equivalents)



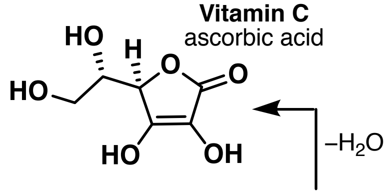
Vitamin A_1
retinol



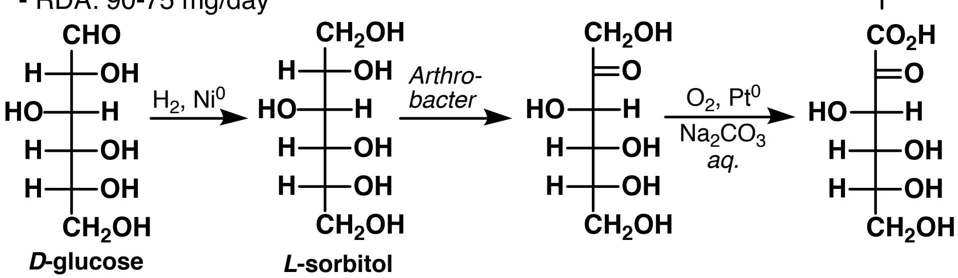
[from citral]

Vitamin C (ascorbic acid):

- strongly reducing properties (reductone)
- isolation done w/o light, oxygen, or copper
- cooking destroys most of it
- commercially syntehsized since 1930's
- RDA: 90-75 mg/day

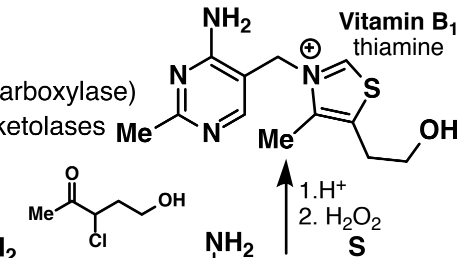


Vitamin C
ascorbic acid

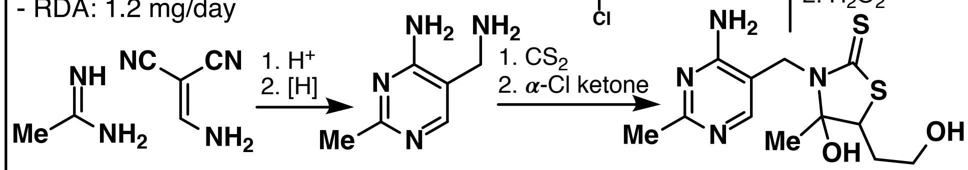


Vitamin B_1 (thiamine):

- first substance called a "vitamin"
- diphosphorylated to become active (cocarboxylase)
- is a cofactor for carboxylases and trasnketolases
- Not present in certain grains (rice)
- oxidatively and reductively labile
- RDA: 1.2 mg/day

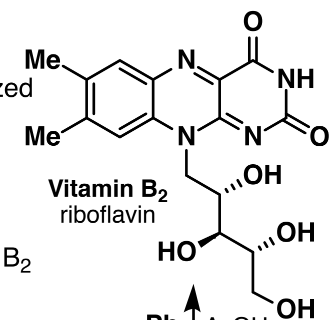


Vitamin B_1
thiamine

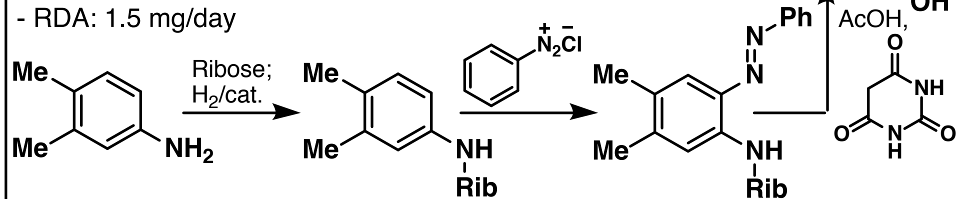


Vitamin B_2 (riboflavin):

- resistant to oxidation, not reduction; easily re-oxidized
- central to oxidase enzymes (FAD- and FMN dependent)
- found in all plants/animals; high concentrations in yeast, livers, and hearts
- numerous countries require fotification of grains w/ B_2
- produced both chemically and by fermentation
- RDA: 1.5 mg/day



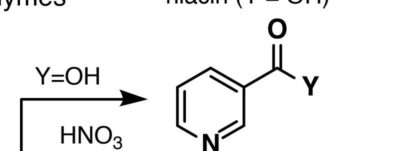
Vitamin B_2
riboflavin

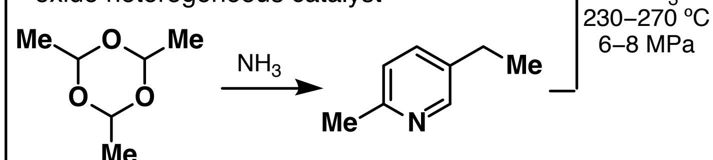


Vitamin B_3 (niacin):

- biologically active as nicotinamide
- becomes part of NAD/NADP-dependent eznymes
- body can convert tryptophan to niacin
- RDA: 15 mg/day
- second process uses Ti/Sb/V or Ti/Sb/U oxide heterogeneous catalyst

Vitamin B_3
nicotinamide (Y = NH_2)
niacin (Y = OH)





- dicarboxylate interm.
- neutralized with S.M.
- NO oxidized to NO_2; water to get HNO_3

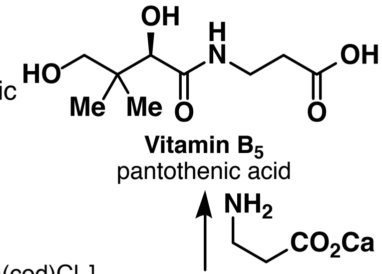
David Peters

Food Chemistry: An Organic Perspective

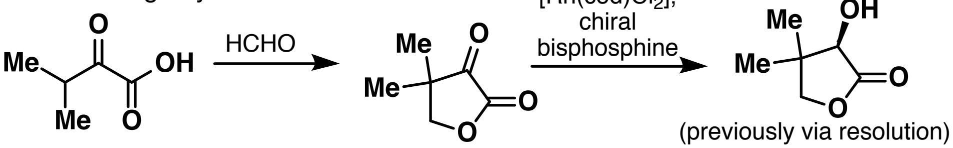
Baran Group Meeting
10/5/19

Vitamin B_5 (pantothenic acid):

- coenzme A precursor
- central to metabolism: fatty acid synthesis, citric acid cycle, acetylation, porphyrin snyth, degredation of fats and carbohydrates, etc.
- deficiency leads to a variety of symptoms
- 80% of world production used in animal feed
- RDA: 5 mg/day

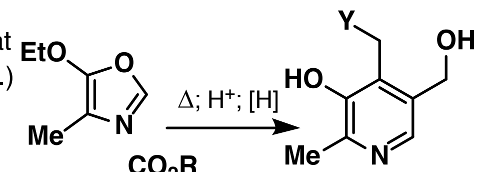


Vitamin B_5
pantothenic acid



Vitamin B_6 (pyridoxine):

- refers to 6 related compounds (those at right and their 5-phosphomethyl derivs.)
- active form as CHO bound in enzyme
- cofactor for transaminase, epimerase, and decarboxylase enzymes
- obtained from eating plants
- Most stable form is pyridoxine HCl
- RDA: 1.3 mg/day

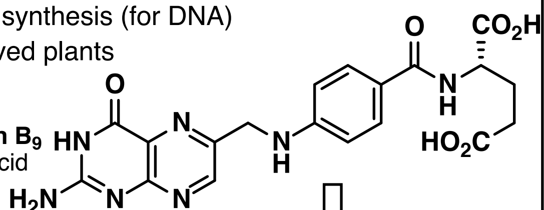


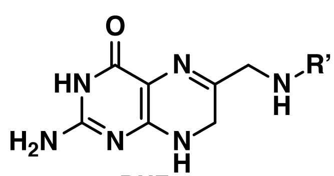
Vitamin B_6
pyridoxine (Y = OH)
pyridoxal (Y = CHO)
pyridoxamine (Y = NH_2)

Vitamin B_9 (folic acid):

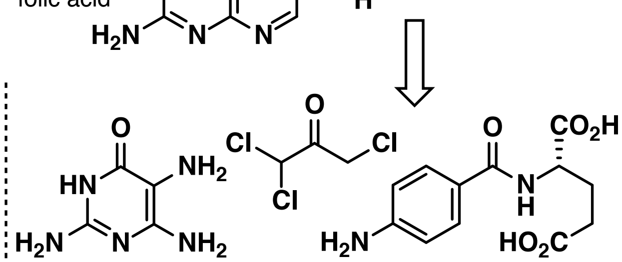
- Dihydro- and tetrahydrofolate are coenzymes for methyl and formyl transfer
- central to methionine and purine synthesis (for DNA)
- high concentrations in green-leaved plants
- lack of folate results in anemia
- metabolic link between vitamins B_3, B_9, and B_{12}
- RDA: 400 µg/day

Vitamin B_9
folic acid



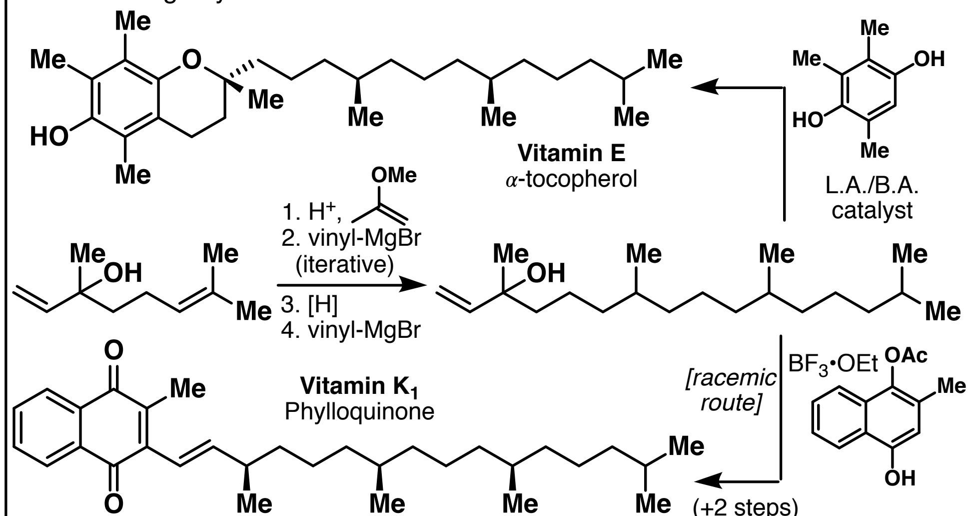


DHF



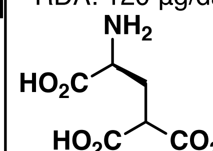
Vitamin E (tocopherols):

- vary in methylation of arene and olefins in tail (tocotrienols)
- antioxidants: delay the oxidation of membrane lipids = increased integrity
- high concentrations in plant oils
- provided usually as all racemic & acetate (~67% activity of natural)
- RDA: 15 mg/day

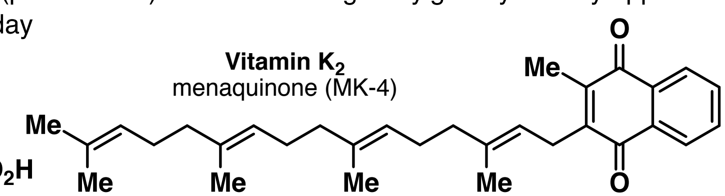


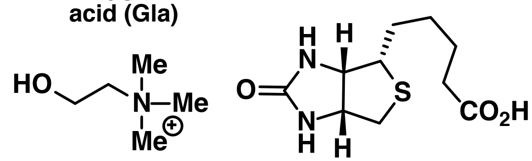
Vitamin K (menaquinones):

- group of related compounds that vary in chain length and unsaturation
- cofactor in glutamic acid carboxylase (Gla synthesis)
- important for blood clotting (Gla binds Ca^{2+} ions)
- phylloquinone (plant-based) is cleaved and geranylgeranyl moiety appended
- RDA: 120 µg/day



carboxyglutamic acid (Gla)





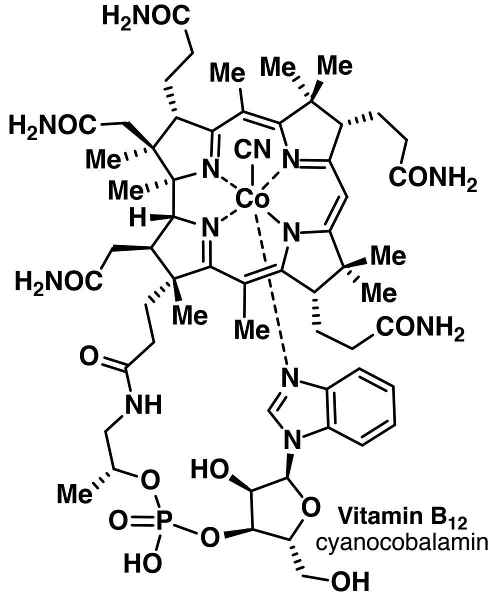
Choline
(Vitamin B_4)

Biotin
(Vitamin B_7)

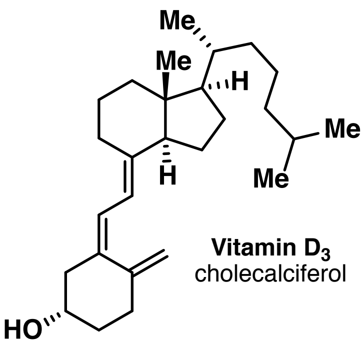
<u>Note on Nomenclature</u>

- leters and numbers are skipped due to reclassification
- biotin and choline have flip flopped (both vitamins now)
- B class are all water soluble & cofactors

Vitamin B_{12} (cobalamins):
- the cobalamins are grouped; vary by axial ligands
- biosynthesized by microorganisms
- most comes from eating meat
- methionine and Suc-CoA synthesis
- humans have a specific mucoprotien for absorbing
- produced by fermentation (*P. dentrificans* - highly mutated strains)
- converted to cyanocbalamin w/ KCN; extracted w/ cresol & recrystalized
- for synthesis see: <u>Classics in Total Synthesis</u>, Chapter 8
- RDA: 2.4 μg/day



Vitamin B_{12}
cyanocobalamin



Vitamin D_3
cholecalciferol

Vitamin D (calciferols):
- central to maintaining calcium and phosphorous homeostasis in body
- converted to active form in body
- In depth discussion of Vitamin D see: *Synthesis of Vitamin D*, Gu, Baran Lab GM
- RDA: 20 μg/day

Minerals:
- nutritional minerals are essential dietary components that are elemental
- does not include C, H, O, N
- some of these fall under macrominerals (Ca, K Mg, Na, P)
- the rest are minor (S, Fe, Cl, Zn, I, Cu, Mn, Mo, Se, Cr, Co)
- others are recognized as important but the roles and levels necessary are unestablished (Br, Li, Ni, F, etc.)

Mineral	RDA (mg)
K	4700
Cl	2300
Na	1500
Ca	1200
P	700
Mg	420
Fe	18
Zn	11
Mn	2.3
Cu	0.9
I	0.15
Cr	0.035
Mo	0.045
Se	0.055
Co	N/A

Other Common Food Additives:

<u>Carbohydrate-based thickeners:</u>
- corn starch
- tapioca starch
- agar
- pectin
- dextrin
- sodium alginate

<u>pH regulators:</u>
- ascorbic acid
- citric acid
- benzoic acid
- sodium benzoate

<u>Anticaking agents:</u>
- silicon dioxide
- Na/Ca silicate
- stearic acid/stearates
- calcium phosphate

<u>Preservatives:</u>
- nitrates
- nitrites
- benzoates
- propionates
- sulfates
- ascorbates
- BHT
- gallic acid