The physical and chemical structure of asphalt: with a brief history of their usage and availability

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Outline

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1. What are Bitumen and Asphalt?
2. On the Abundance of Bitumen in Nature
3. Physical Chemistry of Asphalt

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3. Physical Chemistry of Asphalt

The Dictionary Definition

- Asphalt: A bituminous substance, found in many parts of the world, a smooth, hard, brittle, black or brownish-black resinous mineral, consisting of a mixture of different hydrocarbons; called also mineral pitch, Jew's pitch and in the Old Testament 'slime'

Oxford English Dictionary

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- Bitumen: Originally, a kind of mineral pitch found in Palestine and Babylon, used as mortar, etc. The same as asphalt, mineral pitch, Jew's pitch, Bitumen Judaicum.
- In modern scientific use, the generic name of certain inflammable substances, native hydrocarbons more or less oxygenated, liquid, semi-solid and solid, including naphtha, petroleum, asphalt, etc. Elastic Bitumen: Mineral Caoutchouc or Elaterite.

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The ASTM and the RILEM Definition

- Bituminous materials: a class of black or dark-colored (solid, semi-solid or viscous) cementitious substances, natural or manufactured, composed principally of high molecular weight hydrocarbons, of which asphalts, tars, pitches, and asphaltites are typical.

ASTM D8-97-2000

- Asphalt: A natural or artificial mixture of bitumen with mineral matter

  Bitumen: The heaviest fraction of petroleum; it can be *petroleum bitumen* or *natural bitumen*

RILEM Technical Committees, 1992 (Organic Binders)

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Highway Engineer Vs. Petroleum Chemist

- Asphalt is a substance that causes particles of sand, gravel and crushed stone to stick together to form a pavement.

Highway Engineer

- (Straight run) Asphalt (with the exception of natural asphalt) is a residue of petroleum or a product of solvent extraction of petroleum. It is a complex mixture of thousands of different organic compounds mutually dissolved or dispersed.

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- The word bitumen may, therefore, be strictly defined as a generic term that is used to designate a class of minerals as they occur in nature, ...
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Various Forms of Bitumen

- Bitumen, asphalt, resin, tar, pitch, wax . . .
- Natural Bitumen:
  1. Bitumens with inorganic impurities - Found in lakes in Trinidad, Venezuela, Cuba and as Tar sands in Athabasca etc.

FIG. 6—PITCH LAKE AT TRINIDAD, W. I.—From an old print.

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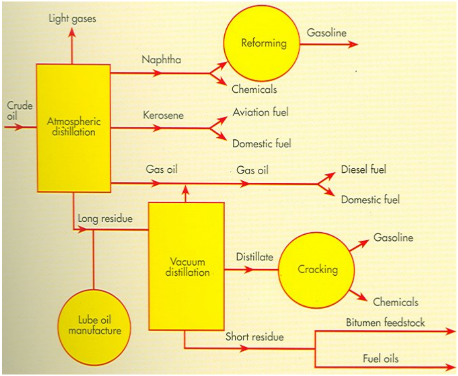

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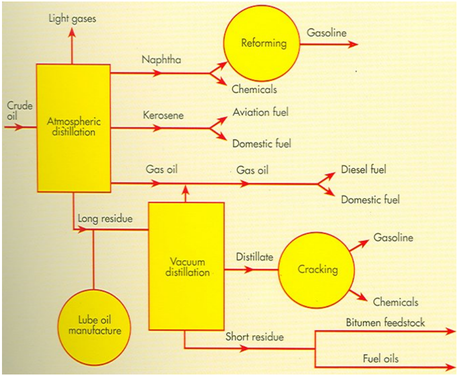

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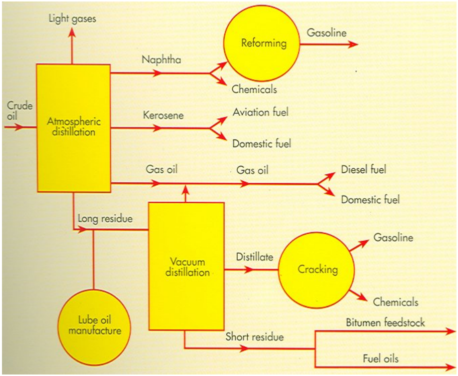

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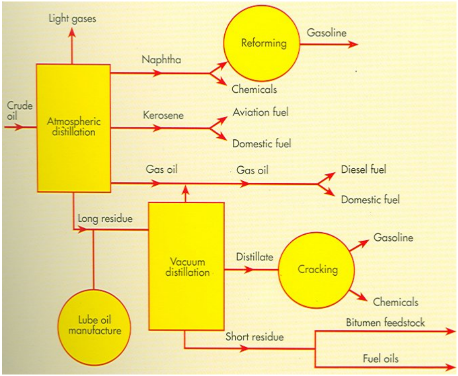

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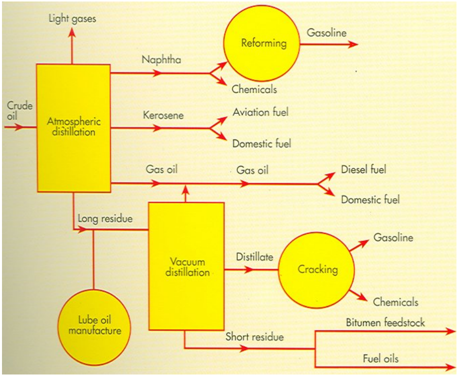

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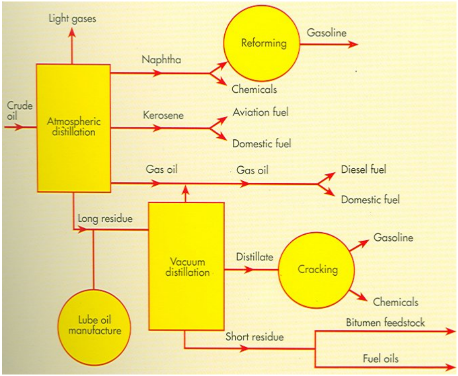

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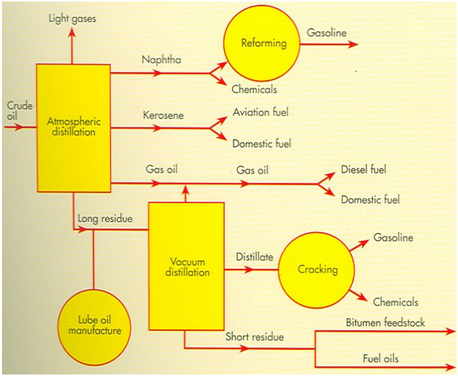
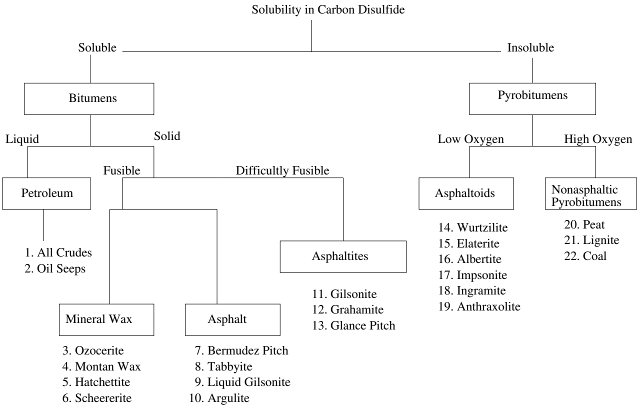


Bitumen Classification Based on Solubility

- Current classification system for bitumen (asphalt): Abraham (1912), Pfeiffer (1950), Chilingarian and Yen (1978)



The Death Trap of Rancho-la-Brea, California

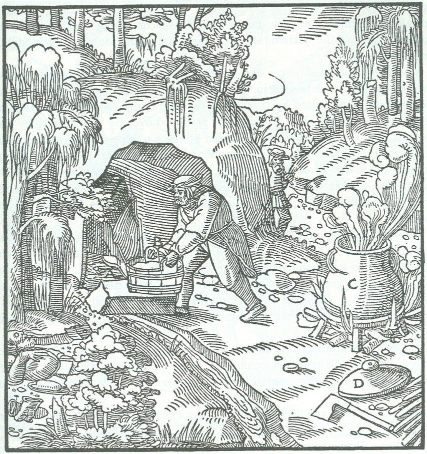
FIG. 1.—"The Death Trap of Rancho-la-Brea, California."
Painted by Charles R. Knight.

- *The pits were originally formed by 'blow-outs' . . . filled by an inflow of soft, sticky 'asphalt', which in time became quiescent, possibly crusting over, but deadly to any form of beast that stepped into them. Once mired in the asphalt, the victim's struggles would sink it deeper and attract a host of carnivores to the feast.*

Agricola's Observation

Liquid bitumen, if there is much floating on springs, streams and rivers, is drawn up in buckets or other vessels; but, if there is little, it is collected with goose wings, pieces of linen, ralla, shreds of reeds and other things to which it easily adheres, and it is boiled in large brass or iron pots by fire and condensed.

Agricola (1556), De Re Metallica



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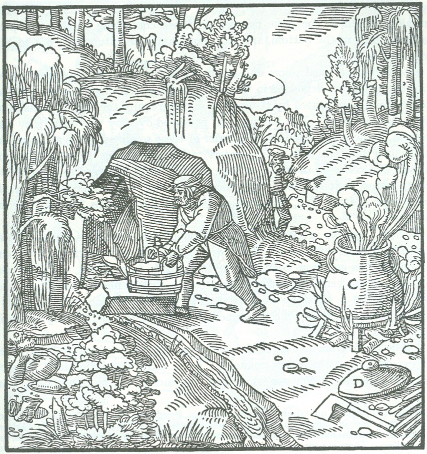

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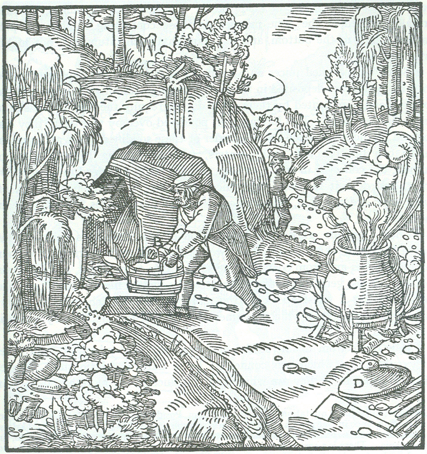

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Bitumen in Dead Sea

"…of this kind is the lake which the Hebrews call the Dead Sea, and which is quite full of bituminous fluids"

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Use of Bitumen in Antiquity - Adhesive

- Adhesive of choice to fix flint to wooden handles

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- A scraper and a Levallios flake from Syria (used around 40,000 BC)

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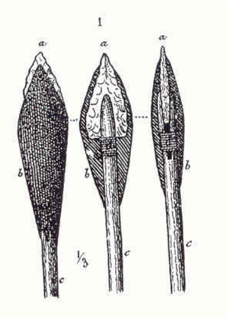

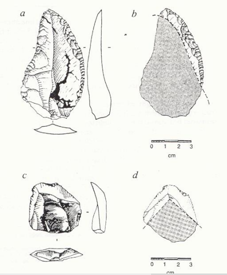

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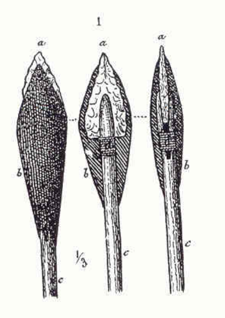

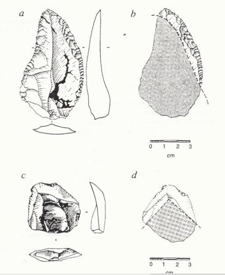

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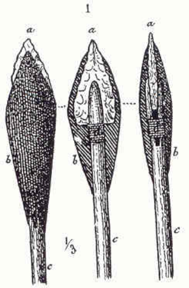

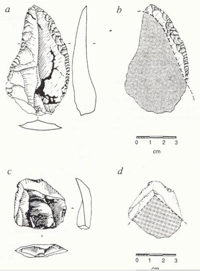

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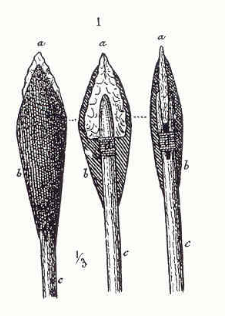

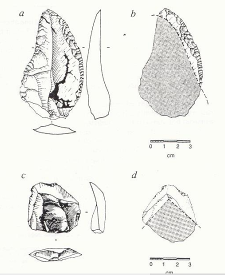

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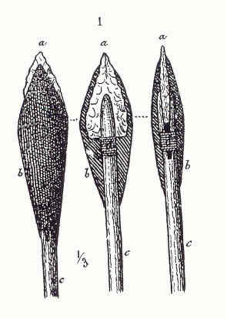

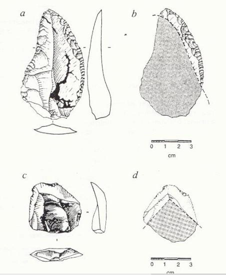

Use of Bitumen in Antiquity

- Main use of bitumen in antiquity and prehistory (Hummalian period - 180,000 BC)

use of bitumen	examples	excavations with examples studied
mortars in construction building	temples, palaces, terraces, floors, ziggurats, door threshold, courtyard	Mari, Babylon, Larsa, Haradum, Qal'at al-Bahrain, Mleiha, Failaka
waterproofing agent	mats, baskets, jars, water reserves, bathrooms, water pipes, cisterns, boats, sarcophagi	Tell es-Sawwan, Tell el'Oueili, Qal'at al-Bahrain, Saar, Baghdad, Ra's al-Junayz, Susa, Failaka, Tell Brak
adhesive and glue	sickles, tool handles, statues, jars, decoration (game, lyre, temple, pillar, ostrich egg)	Tell Atij, Netiv Hagdud, Umm El Tlel, Mari, Tell Halula, Ras Shamra, Susa
domestic artefacts	spindle whorls, balls, dice, wall cones	Tell el'Oueili, Failaka, Saar?, Qal'at al-Bahrain, Susa, Tell Brak
jewellery	bead, ring, gold badges on clothing or for horse harnesses	Umm al-Qaiwwain, Ulu Burun, Susa, Saar
sculpture	sculpture, cylinder and stamp seal of Susa in bitumen mastic	Susa
mummification	mixed with conifer resin, beeswax, grease to prepare mixtures for embalming	Egyptian mummies from the Queen valley and from several Museums (Lyon, Hannover, Paris)

Connan, Phil.Trans. R. Soc. Lond. B (1999) 354, 33–50

Uses of Bitumen from Prehistoric to Modern times - Sculpture

- Bituminous Mastic sculpture at Susa
  - Susa is located in the southwestern Iran, in the Khuzistan province.

  - Sculptures of this form date to 4000 BC were made using Bitumen Mastic (bitumen mixed with varied mineral elements and subjected to a thermal process around 250 °C, an *annealed bitumen mastic*)

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- *. . . a thin wash of bitumen acting as a glue to fix the metal on to the wood. The head and legs were mortised into a rudimentary wooden body which was next rounded off into proper shape with plaster of paris and given a thick coat of bitumen, a thin silver plate was fixed over the belly, and into the bitumen covering back and sides were pressed the locks of hair . . .*

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- The artificial mixture (spices, resins and bitumen) prepared by the Egyptians used to preserve their dead was also called *mumiya*, and a body preserved in this manner was termed *mumiyya* and later in French as *momie* and in English as *mummy*.

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More than 250 Current Known Uses of Bitumen

Agriculture Damp-proofing and water-proofing buildings, structures, Disinfectants, Fence post coating, Mulches, Mulching paper, Paved barn floors, barnyards, feed platforms, etc., Protection tanks, vats, etc., Protection for concrete structures, Tree paints, Water and moisture barriers, (above and below ground) Wind and water erosion control, Weather modification areas. **Buildings** *Floors* Damp-proofing and water proofing, Floor compositions, tiles, coverings, Insulating fabrics,	*Miscellaneous* Air drying paints, varnishes, Artificial timber, Ebonized timber, Insulating paints, Plumbing, pipes, Treated awnings. **Hydraulic and erosion control** Hydraulic and erosion control Canal linings, sealants, Catchment area, basins, Dam groutings, Dam linings, protection, Dike protection, Ditch linings, Drainage gutters, structures, Embankment protection, Groynes, Jetties, Levee protection, Mattresses for levee and bank protection, Membrane linings, water proofing, Reservoir Revetments, Sand dune stabilization, Sewage lagoons, oxidation ponds, Swimming pools,	*Compositions* Black grease, Buffing compounds, Cable splicing compound, Coffin linings, Embalming, Etching compositions, Extenders, Explosives, Fire extinguisher compounds, Joint fillers, Lap cement, Lubricating grease, Pipe coatings, dips, joint seals, Plastic cements, Plasticisers, Preservatives, Printing inks, Well drilling fluid, Wooden cask liners. *Impregnated, treated materials* Armoured bituminized fabrics, Burlap impregnation, Canvas treating, Carpeting medium, Deck cloth impregnation, Fabrics, felts, Mildew prevention, Packing papers, Pipes and pipe wrapping, Planks, Rugs, asphalt base,	**Paving** (See also Hydraulics, Agriculture, Railways, Recreation) Airport runways, taxiways, aprons,etc. Asphalt blocks, Brick fillers, Bridge deck surfacing, Crack fillers, Curbs, gutters, drainage ditches, Floors for buildings, Warehouses, garages, etc. Highways, roads, streets, shoulders, Parking lots, driveways, Portland cement concrete underseal, Roof-deck parking, Sidewalks, footpaths, soil stabilisation.

More than 250 Current known Uses of Bitumen - Continued

papers, Step treads. *Roofing* Building papers, Built-up roof adhesives, felts, primers, Caulking Compounds, Cement waterproofing compounds, Cleats for roofing, Glass wool compositions, Insulating fabrics, felts, papers, Joint filler compounds, Laminated roofing shingles, Liquid-roof coatings, Plastic cements,Shingles. *Walls, siding, ceilings* Acoustical blocks, compositions, felts,Architectural decoration, Bricks, Brick siding, Building blocks, papers, Damp-proofing coatings, compositions, Insulating board, fabrics, felts, paper, Joint filler compounds, Masonry coatings, coatings, Plaster boards, Putty, Siding Compositions, Soundproofing, Stucco base, Wallboard.	Waste ponds, Water barriers. **Industrial** *Aluminum foil compositions using bitumen* Backed felts, Conduit insulation, lamination,Insulating Boards, Paint Compositions, Papers, Pipe wrappings, Roofing, Shingles. *Automotive* Acoustical compositions, felts, Brake linings, Clutch facings, Floor sound deadeners, Friction elements, Insulating felts, Panel Boards, Shim strips, Tacking strips, Underseal. *Electrical* Armature carbons, windings, Battery boxes, carbons, Electrical insulating compounds, papers, tapes, wire coatings, Junction box compound, Moulded Conduits.	Sawdust, cork, asphalt composition, Treated leather, Wrapping papers. *Paints, Varnishes, etc* Acid-proof enamels, mastics, varnishes, Acid-resistant coatings, Air-drying paints, varnishes, Anti-corrosive and anti-fouling paints, Anti-oxidants and solvents, Bases for solvent compositions, Baking and heat resistant enamels, Boat deck sealing compound, Lacquers, japans, Marine enamels *Miscellaneous* Belting, Blasting fuses, Briquette binders, Burial vaults, Casting moulds, Clay articles, Clay pigeons, Depilatory, Expansion joints, Flower pots, Foundry cores, Friction tape, Fuel, Gaskets, Gramophone records, Mirror backing, Rubbers, moulded compositions, Shoe fillers, soles, Table tops.	**Railways** Ballast treatment, Curve lubricant, Dust laying, Paved ballast, sub-ballast, Paved crossings, Freight yards, Station platforms, Rail fillers, Railway sleepers, Sleeper impregnating, stabilisation. **Recreation** *Paved surfaces for:* Dance pavilions, Drive-in movies, Gymnasiums, sport arenas, Playground, school yards, Race tracks, Running tracks, Skating tracks, Swimming and wading pools, Tennis courts, handball courts. *Bases for:* Synthetic playing field and running track surfaces

Uses of Bitumen from Prehistoric to Modern times - Pavement Construction

- 2800 BC - In Asia Minor, Mesopotamia and Persia - Extensive use of Bitumen tracks.
- 2400 BC - India - Extensive use of bitumen joined bricks for pavement.
- 100 BC - North Western Europe - Roman log roads
- 1837 AD - Paris - Seyssel mastic for footpaths
- 1869 AD - London - Asphalt
- 1872 AD - Union Square, New York - Compressed Neuchatel rock asphalt
- 1876 AD - Berlin - Val de Travers asphalt
- 1876 AD - Pennsylvania Avenue, Washington - Rock asphalt, later repaired with Trinidad asphalt mixture

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Uses of Bitumen from Prehistoric to Modern times - Pavements

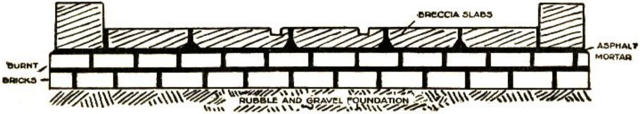


Fig. 10.
Processional road in Temple of Ishtar at Assur (1 : 40).

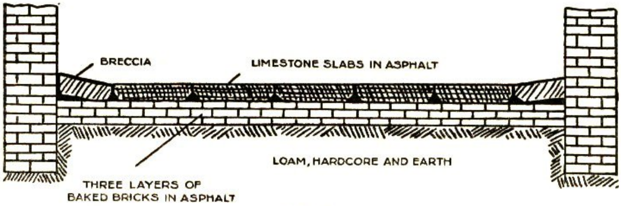
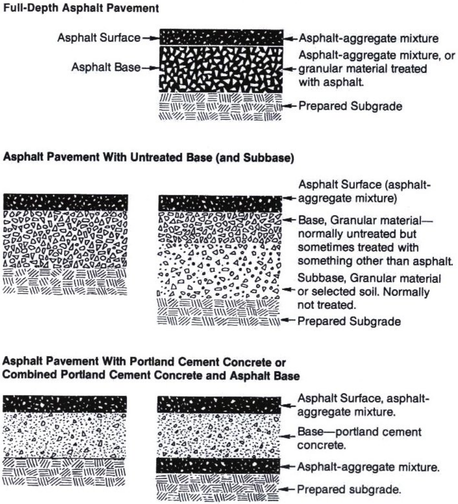


Fig. 11.
Processional road "Aiburshabu" in Babylon (1 : 60).

Uses of Bitumen from Prehistoric to Modern times - A typical pavement cross-section used now



Uses of Bitumen from Prehistoric to Modern times - Bituminization of Radioactive Wastes

- For the past 30 years, more than 20 member Nations of IAEC have been using bituminization of radioactive wastes.
- Nuclear wastes are embedded in molten bitumen (180 to 200 °C), cooled and then disposed off.
- Production of radiolytic gases (H_2, CH_4, CO_2) depends upon the type of bitumen, dosage rate etc.
- Release of radiolytic gases can result in crack formation in oxidized asphalt and bubbling in unoxidized asphalt.
- IAEC uses a combination of empirical tests such as Penetration test, Ring and Ball test and Ductility test for specifications. (*These tests are gradually being phased out in SUPERPAVE specifications for Highway Constructions!!!)*.

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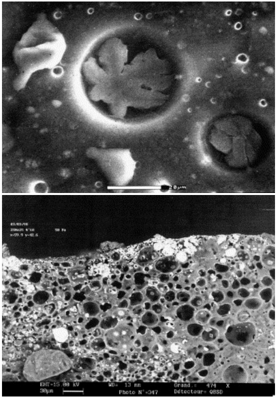
Uses of Bitumen from Prehistoric to Modern times - Bituminized radioactive waste leaching



- Current state of art: Diffusion of water through bitumen - Fick's Law

Constitution of Asphalt

- Boussingault(1837) carried out some of the early experiments on the composition of bitumen (bitumen of Becherlbronn) and classified it into two groups, namely *petrolene* and *asphaltene*.
- *"In conclusion, it is seen that the glutinous bitumens may be considered as mixtures, probably in all proportions, of two principles, each of which has a definite composition. One of these principles (asphaltene) fixed and solid, approaches asphalt in its nature. The other (petrolene) liquid, oily and volatile, resembles in some of its properties, certain varieties of petroleum. It may, then, be conceived that whilst the consistency of bitumen varies, it may be said to infinity; it suffices that one or the other of the two principles dominates the mixture, thereby giving such or such a degree of fluidity."*

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- Kayser (1879) isolated asphalt into 'α', 'β' and 'γ' asphalts *"...α' asphalt - oily, the 'β' asphalt was a solid gummy substance melting at 60 °C and the 'γ' asphalt had the same consistency as 'β' asphalt but melted at 165 °C.*
- Nellensteyn (1924) - *"Asphalt when solidifying shows a very marked increase in viscosity without crystallization. Other bitumens show either crystallisation or a general increase in viscosity."*
  "...asphalt contains elementary carbon in colloidal form and that this colloidal form is the essential constituent of asphalt"
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- Nellensteyn (1931)- Application of colloidal chemistry to investigate asphalt
  - the medium,
  - a lyophile part:the protective bodies and
  - a lyophobe part: the ultramicrons.
- *“The stability of the whole system in the first place depends upon the relation between the micelles and the medium. Changes in this stability, which are known as flocculate and peptizing reactions, give rise to a ‘reversible flocculation’. ”.*
- *“. . . it is certainly very mysterious that the coagulated bitumen easily expels the water, while molten bitumen adheres very poorly to moistened surfaces of mineral matter”.*

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- Asphaltenes, asphaltic resins and oils
- Asphaltenes imparting hardness and high melting point to the mixture
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- Oily constituents acting as the dispersing phase

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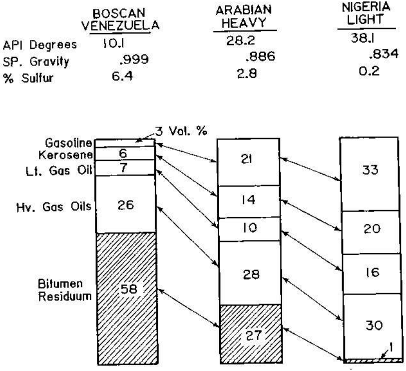
Constitution of Asphalt - First Statement About the Influence of the Crude Source

- Baskin (1932) : *“Asphalt is in no sense a distinct group by itself, but rather a portion of the crude petroleum containing several groups in form of a mixture or mutual solution. The character and makeup of each group to start with depends to a great extent on source. For any given consistency, the oily constituents in a California crude residue are different in physical and chemical properties from those extracted from the same consistency Panuco residual. This likewise, undoubtedly holds true of the resins and the asphaltenes”*

And finally the Corbett Fractions

- Make-up of crude petroleum, Corbett, 1984



And finally the Corbett Fractions

- Summary of composition and characterization as found in typical 85/100 straight reduced asphalt (Corbett:1969)

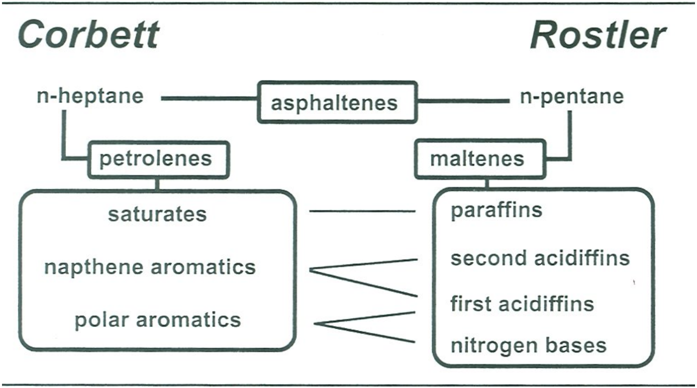
Component	Wt % range	Physical Nature	Density 20/4 °	Molecular Wt (Av)
Saturates	5-15	Colorless liquid	0.87	650
Naphthene-Aromatics	30-45	Yellow to Red Liquid	0.99	725
Polar-Aromatics	30-45	Black Solid	1.07	1150
Asphaltenes	5-20	Brown to Black Solid	1.15	3500

Influence of Crude Source on Corbett Fractions

- Effect of crude source on composition (Corbett:1969)

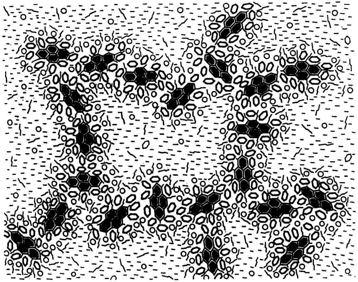
	Venezuelan	USA	Mexico	Mid-East
Saturates	14.0 %	10.5 %	8.5 %	8.0 %
Naphthene-Aromatics	34.5 %	38.5%	29.8 %	38.5 %
Polar-Aromatics	36.3 %	33.4%	42.6 %	37.0 %
Asphaltenes	14.1 %	16.8 %	28.3 %	15.5 %
Penetration @ 77 $^{\circ}$F	90	92	88	85
Soft Point, $^{\circ}$F	114	114	116	115

Corbett Vs. Rostler



Some ‘Structural Models’ for Asphalt

- Pfeiffer and Saal (1940): Asphalts are visualized as colloidal system, with asphaltenes forming the centers of micelle and having a more pronounced aromatic nature, the asphaltenes were assumed to be surrounded by lighter constituents of less aromatic nature, and there were no distinct interphases between the micelles and the medium surrounding it



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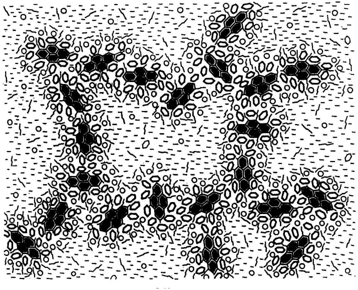

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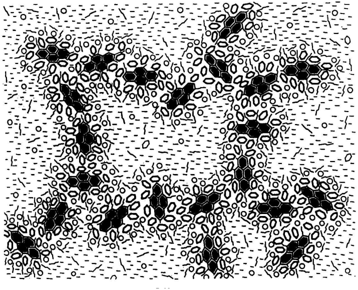

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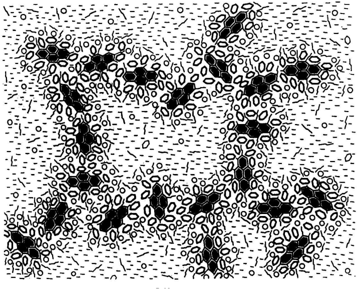

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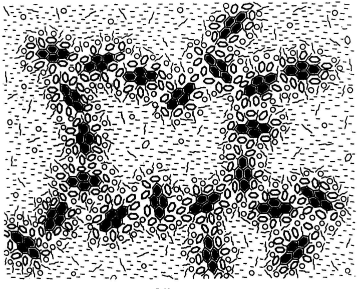

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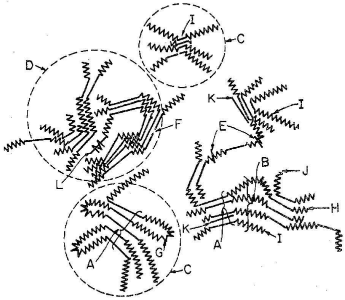

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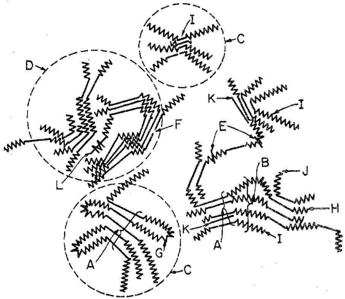

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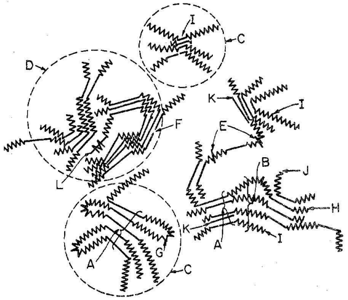

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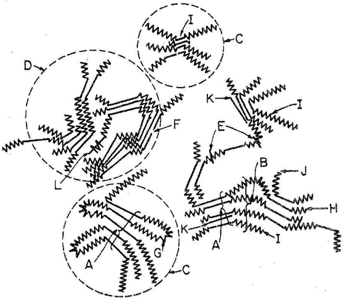

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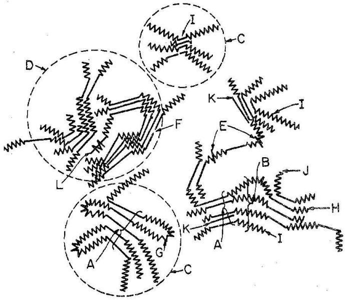

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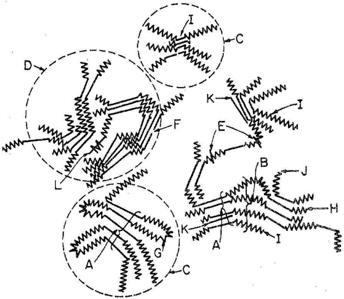

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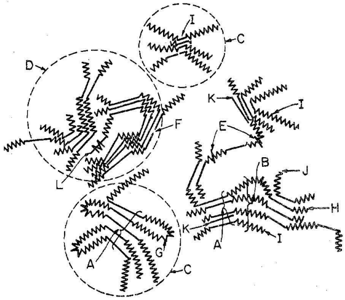

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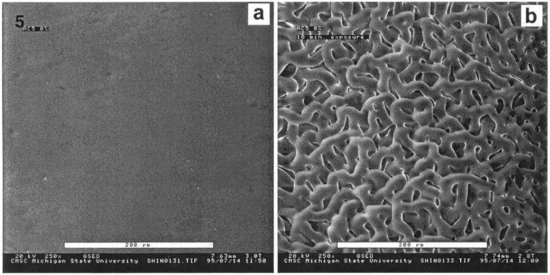

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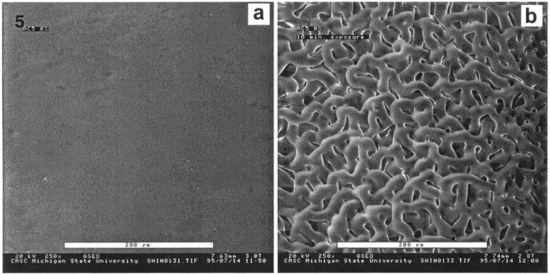

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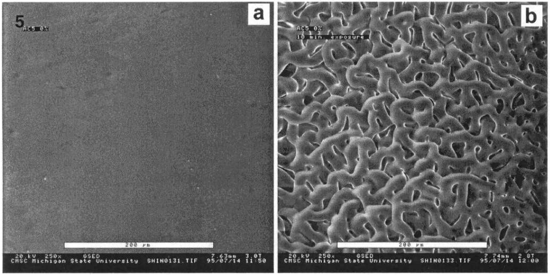

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Multitude of Tests for Unraveling the Structure of Asphalt

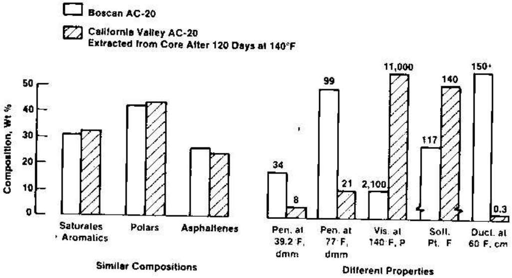
- solvent precipitation,
- chemical precipitation,
- adsorption liquid chromatography,
- ion exchange liquid chromatography,
- coordination liquid chromatography,
- thin-layer chromatography,
- gas-liquid chromatography,
- size-exclusion chromatography,
- high pressure gel permeation chromatography,
- vapor pressure osmometry,
- mass spectrometry,
- electrophotometric spectroscopy,
- nuclear magnetic resonance spectroscopy,
- electron spin resonance spectroscopy,
- spectrochemical analysis,
- elemental analysis,
- distillation fractionation,
- wax content determination,
- photochemical reactions of asphalt,
- acid number determination,
- internal dispersion stability,
- titrimetric/gravimetric analysis . . .

Chemical Composition Vs. Physical Properties

- Goodrich *et al.*(1986):*"Some believe that today's sophisticated analytical tools, computer-controlled instruments that work with milligrams of asphalt, should make the connection between asphalt composition and performance properties. These instruments are being applied to asphalt research and well-defined field problems. Fundamental chemical explanations of asphalt aging, adhesion, structure, and rheology have been proposed. Yet the complex chemical mix of even a single asphalt may never be adequately described."*

Chemical Composition Vs. Physical Properties

- Asphalts with similar compositions but different properties from Goodrich *et al.*(1986)



The SHRP Initiative of USA

- Arab Oil Embargo in 1970’s resulted in United States processing crudes from different refineries.
- “Asphaltenes: Where are you?”
- “The asphalts are no good nowadays”
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- The SHRP spent a total of $150 million on research on asphalt concrete out of which $50 million was spent on asphalt research on a five year research program that started on 1987.

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  "A major effort has been the study of the rheology, or the viscoelatic properties, to determine the effects of shear, shear rate, and temperature. This effort has resulted in description of asphalt in terms of rheological master curves that show the variation in viscous and elastic components with shear and temperature. In general, all asphalts exhibit a glass like behavior at very low temperature, and are relatively fluid at high temperature, but the pathway from glass to fluid, or vice versa varies substantially from one asphalt to another."

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- Viscosity (Pa·s) of Asphalts and Asphalts blended with IEC fractions at 60 °C1 rad/s from SHRP Report 686, 1994

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- The physical and chemical structure of asphalt is quite complex
- In the next talk, we will discuss a modeling effort that takes into account, some of the complexities of asphalt

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