Fibre Flow Research History: Part I. Background and Beginning

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ABSTRACT

The historic development of fibre flow research reveals that today's direction is the result of a rather limited number of influences; here labelled the technical, the physical, the hydrodynamic, the colloidal and the macro-molecular.

INTRODUCTION

In the normally short introductions to scientific articles there are seldom room for more than a sketchy presentation of the scientific background with a close time horizon. For fibre flow this often means a microhydrodynamic literature review. The actual history is, however, more composed and also more interesting. This work is an attempt to give a fuller presentation of the cultural soil from which this research has grown. This is not only of historic interest but also necessary to understand today's direction, and may therefore also serve as a help to realise that many alternative approaches exist for fibre flow.

It is not possible to write a straight history for such a complex development. Instead, the focus will successively move between the different influences. Interaction between them has, of course, been common, often also in one and the same person.

TECHNICAL TRADITIONS

Hagen-Poiseuille's equation was developed between about 1840 and 1860. The pressure

drop in pipes could then be calculated if the "rheological" material property viscosity was known or could be measured. Before the centrifugal pump was invented in the second half of the 19th century pulp suspensions, then called stuff. were pumped with different types of displacement pumps, e.g. plunger pumps. Knowledge of the fluid properties was not then absolutely essential. It was more a matter of using large enough pipes, pumps and motors to deliver sufficient quantities of stuff. With the introduction of centrifugal pumps the flow properties of the stuff, became an important factor. Trimbey¹ in 1907 seems to have been the first to measure the pressure drop in tubes at different concentrations and velocities.

The first theoretical attempts naturally followed the hydrodynamic path, e.g. Pfarr² and Haussner³ (1907)(1908). The researchers soon realised that the stuff did not behave like ordinary fluids that e.g. could be characterised by a viscosity. The practical problem was met with leaflets and brochures from machine manufacturers, see e.g. Baldwin and van den Akker⁴ (1939). But even when more general solutions were searched, it was not at that time (or is today⁵) self-evident that a material property could be assigned the stuff. And rheology was not yet an established discipline.

The reason behind the strange pipe pressure drops for the stuff was understood

already in the 1920's, *viz.* that the entire pipe cross section was not sheared but that a central plug formed, Forrest and Grierson.⁶ A thorough investigation of the same type was made in the middle of the 1930's by Brecht and Heller^{7,8} [Berthold's brother, paper technology professor in Darmstadt].

The first researcher with a deeper understanding of stuff flow, however seems to have been the Dane Sigurd Smith⁹ (1919), head the laboratories of De Forenede Papirfabrikker A/S. In his thesis work on beating he also studied the open channel flow in a Hollander, where the stuff (5-10%) is repeatedly returned to the tackle and where the actual milling takes place. In this gravity driven open channel flow Smith demonstrated plug flow (a straight line painted on top of the stuff remained straight, and sticks pushed vertically through the stuff remained vertical), *yield* stress (a stationary sloping surface kept its slope, an 11 lb. spanner on top of the stuff did not sink), and he also observed network ruptures ("cleavages"), although he gave no rules for their orientation or the formation mechanism. He even measured the frictional force in the ruptures (with wooden "boats"), etc. Very picturesquely and also appropriately he compared this plug flow to "the valleyward movement of a glacier under the pressure of the upper masses of ice." He explained the plug flow form with that the friction between the wall and stuff was less than within the stuff. Smith's work was regarded as so important by the branch that it was translated to English.¹⁰ In spite of this, its influence on the academic fibre flow research has been negligible. Twenty years later Baldwin and van den Akker⁴ followed up the frictional aspects of Smith's work.

INFLUENCES FROM PHYSICS

It is perhaps difficult to understand today that atoms were not generally accepted as

late as at the beginning of the 20th century. Chemists have used them as practical tools since the late 18th century and already physicists like Gassendi, Hooke and Newton were atomists. Some stalwarts, however, demanded more direct proofs before accepting them as more than possibly a useful working hypothesis. For example Kelvin, who died in 1907, was in later years against almost everything new. [A biographer divided his career in two parts; a first when it was impossible for him to do anything wrong, and a second when it was impossible to get anything right.] He was against atoms (energy was better), he was against Maxwell's equations (his own were better), he did not believe in Darwin (God was better), in 1897 Lanchester's manuscript on aerial flight was refuted since "everyone knows that it is impossible to fly with a machine heavier than air", etc. Another influential opponent of atoms was Wilhelm Ostwald [Nobel Prize¹¹ in 1909, founder and editor of Zeitschrift für physikalische Chemie] who, like Ernst Mach, favoured energy. Ostwald's debates with the atomist Ludwig Boltzmann are legendary, but both were stimulated and remained close friends.

In 1906 Albert Einstein [Nobel Prize 1921] published an article in Annalen der Physik entitled "Eine neue Bestimmung der Moleküldimensionen" containing his thesis work.¹² By solving Navier-Stokes' equation he had obtained the viscosity of a dilute suspension of rigid sphere as $\mu = \mu_0(1 + c)$, where μ_0 is the liquid viscosity and *c* the volumetric content of spheres.

Who does not want to share the glamour (even if just a little) of one of the all-time greatest scientists? Thus, Einstein often thrones at the beginning of texts of suspension flow, although he personally does not seem to have been too engaged in suspension viscosity. As the title indicates, his primary interest instead was to prove just the existence of atoms. Most of his

work during this period fits into this master plan; the photoelectric effect and the Brownian motion,¹³ specific heat, etc. In his thesis work he just applied a solution method in Kirchhoff's textbook from 1877^{14} . In addition he contributed with an error. Later Bancelin,^{15,16} found that his experimental results did not fit with this formula, informed Einstein, who had the calculations checked. The trivial misscalculation was found and the formula was corrected¹⁷ to $\mu = \mu_0(1 + 5c/2)$. [We will return to this illuminating mistake since it has something principal to tell about microhydrodynamics]. Bancelin had used micron-sized gamboge spheres fractionated with a centrifugal technique developed by the physical chemist Jean Perrin¹⁸ at Sorbonne [Nobel Prize in 1926 "for his work on the discontinuous structure of matter, and especially for his discovery of sedimentation equilibrium"]. Perrin (1908) had found that such spheres suspended in a liquid with a just slightly lower density did not sink to the bottom due to bombardment by the liquid molecules, i.e. the Brownian motion [the idea of using sedimentation equilibrium to study Brownian motion came from Smoluchowski, see below]. These systems therefore came into focus in the debate for and against atoms, Perrin.^{19,20}

What was later to be called Brownian motion had in the late 1820's been described by the mineralogist Adolphe Brongniart²¹ in France and the botanist Robert Brown²² in England. The latter had found it in pollen and also demonstrated that it was of nonbiological origin. What it actually was, was at that time not certain. One suggestion was thermal currents caused by microscope illumination, S. Exner (1867).²³ It was not until the late 1880's and when it had been enough convincingly shown, by e.g. Leon Gouy,²⁴ that this ongoing motion was a property of the material itself that it attracted the interest of the physicists. Due to its jerkiness, attempts to measure its

velocity was met with less success, e.g. by Felix Exner²³ (1900) at the University of Vienna, who had inherited his interest from his father S. Exner. Einstein¹³ reformulated problem in terms particle the of displacements (mean square) and developed his theory. The same reformulation had, however, been made earlier but not been published and a similar theory developed by the polish theoretical physicist Marian von Smoluchowski²⁵ in Krakow [but born, raised and educated in Vienna, Joseph Stefan's and Felix Exner's student, 1896-97 in Glasgow at Kelvin's laboratory, 1905-06 at Cavendish with J.J. Thomson, 1916 "Silber Edelweiss" by the German and Austrian Alpine Society]. It was these theories that inspired Perrin to his studies with carefully microscope fractionated spheres. The gamboge agreement with the atom-based theory was so good that even the arch-sceptic Ostwald converted to atomism. An early experimental attempt to verify the theory with the help of colloidal gold and Siedetopf's and Zsigmondi's ultramicroscope had also been made in Uppsala by Theodor (The) Svedberg²⁶ (1906) [Nobel Prize 1926]. Another pioneering experiment was Victor Henri's²⁷ (1908) cinematographic studies of Brownian motion in microscope.

COLLOIDAL INFLUENCES

Perrin's gamboge sphere studies may be viewed as a part of the then very active *colloid science*. The colloidal gold system was at that time studied by e.g. Svedberg^{26,28} (1906, 1912) with ultracentrifuges and by Richard Zsigmondy [Nobel Prize 1925] with ultra-microscope. A scientist of the previous generation who took part in the debate was Svante Arrhenius [Nobel Prize 1903]. Already in 1887 he had been interested in the molecular reason behind the viscosity of solutions²⁹ and had in 1915 extended his interest to the viscosity of colloids³⁰ (sulphur, proteins, globulins, egg albumin, etc). His logarithmic correlation was better than Einstein's over a wider concentration range, but had a more general scientific character than microhydrodynamics.

The mechanism behind the coagulation of colloidal systems was at that time not at all clear. A theory did not come by itself, Zsigmondy and had to encourage Smoluchowski to work with these systems. Smoluchowski, with interests also in microhydrodynamics,^{31,32} identified Brownian motion as the key factor. With this approach, a solute molecule differs from a colloidal particle only in mass. He classified the suspended colloidal particles in singlets, doublets, triplets, etc., applied Brownian motion theory that implied that singlets were more mobile than doublets, etc. and then applied a statistical kind of reasoning reminiscent of that in Maxwell's and Boltzmann's gas kinetic theories. It is for this conceptual breakthrough^{33,34} in the understanding of the coagulation process that Smoluchowski is remembered. [He died already in 1917 but would otherwise certainly have got the Nobel Prize for it.]

His theory, however, only gave the outlines, so mechanistic details remained to be filled in. In 1916 his theoretical and also experimental interest had turned the viscosity of colloids,³⁵ e.g. the viscosity results for protein solutions obtained by Wolfgang Joseph Pauli³⁶ (1913) [a doctor of medicine who, under the influence of his friend Ernst Mach turned to science and became a biochemistry professor at the Institut für medizinische Kolloidchemie der Universität Wien. Father of the physicist Wolfgang Ernst Pauli, Nobelist in 1945, second name after his godfather Mach, in the literature then naming himself Wolfgang Pauli Jr.].

The colloidal advancements were noticed also in the pulp and paper branch. Thus Campbell and Yorsten³⁷ in 1932 studied the re-flocculation of very dilute fibre suspensions after a mesh, Strachan³⁸ in 1935 discussed the effect on milling of e.g. "colloidially active material" and Wollwage³⁹ in 1939 studied flocculation in laminar tube flow in technically extremely dilute fibre suspensions (0.01%).

HYDRODYNAMIC INFLUENCES

A kind of mechanistic contribution to coagulation came in 1922 when G. K. Jeffery⁴⁰ in Cambridge extended Einstein's calculations to ellipsoids. Jeffery was, however, worried about the fact that the ellipsoids according to his solution continued to move in the same orbits they started in. This e.g. made the viscosity strongly dependent on the initial ellipsoid orientation distribution. Therefore, he suggested that they in reality drifted toward a position of least dissipation rate, by referring to a theorem of von Helmholz and Korteweg, which had, however, been proved only for stationary systems.

Besides this reflection, Jeffery's work, however, clearly belongs to the microhydrodynamic tradition, which goes back to the middle of 19th century when Navier-Stokes' equation (then named Navier-Poisson's eq.) had been well established in 1845⁴¹ and applied for a sedimenting sphere in 1851⁴² by Stokes. As the title of Stokes' impressive memoir indicates, this work was a theoretical sequel to the very important longitude problem of navigation that got its practical solution already in the 18th century by the clockmaker John Harrison.⁴³ Stokes made hydrodynamics "micro" by applying it to e.g. clouds. The development continued with falling ellipsoids bv Oberbeek in 1887, movement in dilational flows by Einstein^{12,17} around 1900, close to walls by Smoluchowski³¹ around 1910 until Jeffery came with his contribution in 1922.

Geoffrey Taylor⁴⁴ (1923) in Cambridge realised that he could test Jeffery's conjecture with his large Couette instru-

ment. The very practical Taylor fabricated tiny aluminium ellipsoids (prolate and oblate, lengths from 1 to 3 mm) with a lathe, placed them in his instrument filled with waterglass (Newtonian). He found that Jeffery's conjecture was correct, but seems to have missed the point of Jeffery's reasoning by trying (in vain) to find an explanation within hydrodynamics, i.e. with Navier-Stokes' equation. This was perhaps not exceptional at that time, since the relevant thermodynamic development came in 1931/32 by Lars Onsager^{45,46} [Nobel Prize 1968], based on micro-reversibility and going back to ideas of Kelvin (then Thomsson⁴⁷) in 1854. Still in 1938 Johannes Burgers⁴⁸ in Delft had problems in understanding the final orientation result from a hydrodynamic point of view, but saved himself by suggesting that, at the molecular level in which he was interested. the Brownian motion effects dominated.

This debate continued well into the 1950's, when Saffman⁴⁹ and Batchelor (Taylor's successor in Cambridge) managed to convince Taylor that his old result had been due viscoelasticity, then popularised by Weissenberg through his striking exeperiments. In 1967 Goldsmith and Mason⁵⁰ suggested that elongated particles sometimes oriented themselves for maximum dissipation rate and sometimes for minimum. Two years earlier Verhás⁵¹ had shown that Navier-Stokes' equation follows from Onsager's principle of least energy dissipation rate for a substance with response described by a viscosity material parameter.⁵²

Returning to Jeffery⁴⁰ he had suggested, to avoid the problems with e.g. the viscosity, to use of an initial random orientational distribution. He gave no mechanistic motivation, but Brownian motion is close at hand. Particle orientation in flow fields was studied both theoretically and experimentally in the late 1920's and early 1930's with suspended material that

became double-refracting upon deformation (kautschuk, polystyrol, gelatine) by Walter Kuhn⁵³⁻⁵⁶ in Karlsruhe, Eugen Guth⁵⁶ and Friedrich Roland Eirich et al.⁵⁷ in Vienna. Particle rotation due to Brownian motion Richard Gans⁵⁸ in was treated by Königsberg and by Eisenschitz⁵⁹ at the Kaiser Wilhelm Institut. [KWI founded in 1911 with industry donations at the centenary jubilee of the University of Berlin, was renamed Max Planck Institut after World War II], see also Burgers,⁴⁸ who further developed this mathematically fairly complicated subject.

A central question for such systems concerns their thixotropy, i.e. why stress falls after the initial stress increase caused by a step increase in deformation rate (and *vice versa*), resulting in pseudoplasticity. Goodeve⁶⁰ in 1939 discussed possible mechanisms, e.g. shear orientation, reduction of particle size by shear, interaction between the particles, etc.

MACROMOLECULAR INFLUENCES

The dust after the battle of atoms had hardly settled when another infected conflict blew up; this time about macromolecules⁶¹. The influential chemist Emil Fischer [Nobelist 1902 for the Fischer-Tropsch method] had declared that organic molecules with a mole weight greater than 5000 grams do not exist. Wolfgang Ostwald, Wilhelm's son, postulated that e.g. starch, cellulose, silk, rubber etc. were colloidal aggregates. X-ray crystallography was proposed 1908 by Max von Laue [Nobel Prize 1914] at KWI in Berlin-Dahlem, and was developed by the Braggs, father and son during World War I [both Nobelists 1915, the latter Cavendish professor in Cambridge]. Such studies, e.g. by Nishikawa and Ono in 1913, did not reveal unit cells larger than those of ordinary molecules, thereby supporting the aggregate theory.

This view was the established when in 1917 Hermann Staudinger [Nobel Prize

1953] declared that such substances instead consisted of giant molecules of covalently linked small-molecule constituents and started campaigning for his idea. Finally, the academic establishment, among them Fritz Haber [Nobel Prize 1918, the Haber-Boschprocess, director of KWI, Berlin-Dahlem], thought that the matter ought to be ventilated at a symposium in Düsseldorf to which Staudinger should be invited. In the background figured personal antipathies between the staunch pacifist Staudinger and Haber. The idea seems to have been to gather Staudinger's opponents against him at the 1926 symposium, but the plan backfired and Staudinger earned more proselvtes than he lost, among them the symposium chairman Richard Willstätter [Nobel Prize in 1915 for the chlorophyll structure].

One speaker was Hermann Mark, employed in 1922 by Haber at the new KWI Institute for Fibre Research. Under Michael Polanyi, he had embarked on X-ray crystallography studies of various natural fibres, e.g. cellulose and silk.⁶² At the Düsseldorf symposium, Mark had not yet made up his mind on the central question. He later developed a compromising theory, the 1930's finally but in accepted Staudinger's idea. Different views, however, developed between them regarding the nature of the macromolecules. Staudinger thought that their backbones were stiff whereas Mark held the view that they were flexible. Here, time was on Mark's side.

After a session as research director at IG Farben, the energetic Mark [called "der Geheimrat" because he was just the opposite, i.e. non-pompous, alpinist and once a member of the Austrian football team] was forced in 1932 for political reasons to give up his post. Hitler's Machtübernahme in 1933 was foreseen and Mark's mother was Jewish. He was instead appointed professor in physical chemistry at the University of Vienna. There he designed world's first curriculum of polymer physics.⁶³ Mark's main interests were polymerisation mechanisms and the viscosity of polymer solutions; a method used by Staudinger⁶⁴ to estimate the molecular weight of the macromolecules, but rather uncertain with а theoretical foundation, see Eisenschitz⁶⁵ and Burgers⁴⁸. For dilute solutions, he had found a linear relationship $\mu_{sp} \equiv \mu/\mu_0 - 1 \propto cM$ between the specific viscosity μ_{sp} , concentration *c*, and the macromolecular weight, using an efficient radius concept (Wirkungsbereich) [cf. the critical concentration and crowding factor reasonings after the 1950's for fibre flow]. This model had to be modified if the macromolecules flexible were (Mark-Houwink equation). Mark's assistant Eirich, who had finished his thesis in 1929 and continued as assistant under his professor Wolfgang Joseph Pauli, carried out the rheological work.

Mark at I. Chemische Laboratorium and Guth at the Institut für theoretische Physik der Universität Wien in 1933 made an inventory of the state of the art and concluded that experimental data were lagging behind the theoretical development.⁶⁶ An interdisciplinary co-operative research was organised, along the same lines as that done by Haber in KWI Berlin-Dahlem [the idea of scientific branch institutes originated from the needs of the textile industry in England in the 19th century, the first being at Shirley in Manchester]. The results were reported between 1936 and 1937 in an impressive series of articles in Kolloid-Zeitschrift under the heading "Untersuchungen über die Viskosität von Suspensionen und Lösungen": Guth^{56,67}, Krasny-Ergen⁶⁸, Guth and Simha⁶⁹, Eirich, Margaretha and Bunzl^{57,70}, Simha⁷¹, Eirich and Goldschmid⁷¹. It covered different aspects such electroviscosity, wall-effects, as the influence of Brownian motion, fibre suspensions, inertial effects, etc. Eirich et al.⁵⁷ also presented cinematic studies of the motion of different types of fibres in various flow fields, *cf.* Henri²⁷. On 13 September 1937, Eirich and Robert Simha⁷³ submitted an article about the interaction between ellipsoids by applying Maxwell's treatment for gases, i.e. all molecules were assumed to be fixed except one that is shot through the assembly. They found that the effective collision cross section (Wirkungs-querschnitt) increased linearly with axis ratios larger than about 2 to 3. This is the first work about a more detailed mechanism of the interaction between two elongated particles known to the author.

Anschluß came on 14 March 1938. Mark, partly because of his friendship with Chancellor Dolfuss who was murdered in 1934 by Nazi conspirators, was dismissed from his professorship and imprisoned by the Gestapo, but he managed to bribe himself free (a years professor's fee) and flee with his family via Switzerland (with the car front draped with swastika and his means hidden as platinum thread in cloth hangers), first to Bragg at the Cavendish Laboratory and then further to Canada.

There he worked at the Canadian International Paper in Hawkesbury for two years on the pulp process, cellulose acetate and viscose. Viscose for tyre cords was the economically most important product for the sulphite pulp industry before nylon, polyester and finally steel took over. The viscose work led to contacts with and later a move to DuPont. There, tyre projects led to contacts with and a move to the Polytechnic Institute of Brooklyn in 1940. In Brooklyn, Mark organised the polymer research that in 1947 was given the name the Polymer Research Institute (PRI). Mark also gathered around him refugee scientists from Europe, among them Eirich.

Eirich became Privatdozent in early 1938 at Mark's department but already in July 19th he was forbidden to lecture because a grandmother was Jewish. He also managed to escape to Cambridge where he worked for some time. When the hostilities started, foreigners, including Eirich, were interned in camps as *enemy aliens*, and Cambridge was declared to be protected area closed to foreigners. In August/September 1940 he was deported on HMS Dunera to Australia, while his wife remained in a camp in England. For two years, he worked at the University of Melbourne on research into explosives⁷⁴ before he was allowed to return to Cambridge in 1944. In 1947 he moved to Mark at PRI, where he continued with polymer research.

In the early 1950's, Eirich was sent to Europe to re-establish contacts and collect information. Mark had many times met and personally knew Börje Steenberg, professor in Paper Technology at the Royal Institute of Technology (KTH) and head of the neighbouring Swedish Paper and Pulp Research Institute (STFI). [His professor Westgren, Physical Chemistry, Arne Stockholm University, at the beginning of century studied colloidal sulphur the cited systems and is frequently in Smoluchowski³³] A seminar was arranged where Eirich spoke about ongoing research at PRI and corresponding information was presented about the activities at KTH/STFI. Amongst other things, a film was shown about fibre motion in shear fields that Steenberg just had got from Stanley Mason, professor in Physical Chemistry at McGill University and also at Paprican/Montreal with whom Steenberg co-operated and exchanged students. During this period he visited America at least once a year, and then normally also went up to Montreal. After the film, Eirich said that this was his experiment, which was a surprise to all since this was the latest from Montreal.

Eirich then described his pre-war research in Vienna, and told that his rheological involvement had started with haemorheology in co-operation with the central Hospital in Vienna, and then continued with kaolin suspensions to model the stacking of the red blood cells. When, in 1938, he was forced to flee over the Alps, he had time only to collect a small film roll with a content similar to that just shown. When he then was deported to Australia, the film box was left behind on a bench at Cavendish. Since his European tour would now also take him to England and he would afterwards return to his base at STFI, he went up to Cambridge and found the film on exactly the same spot where it had been left. When the film box was opened for the first time at KTH it was found that it had fragmented and could not be shown. At KTH there was then a professor in photography who had managed to save the undeveloped film rolls after 30 years in the Arctic after the unsuccessful attempt to reach the North Pole in balloon by Salomon August Andrée in 1897 [and which ended in tragedy]. This professor restored Eirich's film, which confirmed what he had said. This film was mixed with Mason's film and regularly shown in the course in Paper Technology at KTH during Steenberg's time as professor.

Then it was neglected. A year ago the author, with the ambition to transfer it to CD, managed to identify it in a large heap of unmarked film rolls at the department. Last summer he and Steenberg held the roll in their hands and looked at the frames and read the German text against the daylight because the film projector lacked a lens. But before a lens had been borrowed, the film suddenly was gone when a person left the department. So now, the film that survived Anschluß, flight over the Alps, Blitzen, fragmentation at Cavendish, restoration at KTH is gone again! Will it resurface again, or is it now forever lost?

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