

UNITED STATES PATENT AND TRADEMARK OFFICE

---

BEFORE THE PATENT TRIAL AND APPEAL BOARD

---

BAKER HUGHES INCORPORATED,  
Petitioner

v.

LUBRIZOL SPECIALTY PRODUCTS, INC.,  
Patent Owner

---

IPR2016-01896  
Patent No. 8,022,118

IPR2016-01901  
Patent No. 8,450,249

IPR2016-01903  
Patent No. 8,426,498

IPR2016-01905  
Patent No. 8,450,250

---

**DECLARATION OF PROFESSOR THOMAS H. EPPS, III**

Baker Hughes Incorporated  
**Exhibit 1041**  
BAKER HUGHES v. LUBRIZOL  
IPR2016-01896

## **TABLE OF CONTENTS**

I.	Introduction.....	1
II.	Background and Qualifications .....	2
III.	Documents Considered in Forming My Opinions .....	8
IV.	Person of Ordinary Skill in the Art.....	9
V.	Drag Reducing Polymers.....	11
VI.	The Lubrizol Patents.....	14
VII.	The Holtmyer Publication .....	24
VIII.	The Combination of the Holtmyer Publication and Carnahan .....	37
IX.	The Combination of the Holtmyer Publication and Strausz .....	48
X.	The Inaoka Patent Application Publication.....	62
XI.	The Combination of Inaoka and Carnahan.....	73
XII.	The Combination of Inaoka and Strausz .....	82
XIII.	Conclusion .....	95

I, Professor Thomas H. Epps, III, declare as follows:

**I. Introduction**

1. I have been retained as an expert witness on behalf of Baker Hughes Incorporated (“Baker Hughes”) for the above-captioned *inter partes* review (IPR). I am being compensated for my time at my standard consulting rate, which is \$400 per hour, plus actual expenses. My compensation is not dependent in any way upon the outcome of this matter.

2. I understand that the Baker Hughes petitions for *inter partes* review concern U.S. Patent No. 8,022,118 B2 (“the 118 patent”), U.S. Patent No. 8,426,498 B2 (“the 498 patent”), U.S. Patent No. 8,450,249 (“the 249 patent”), and U.S. Patent 8,450,250 (“the 250 patent”), which I refer to collectively as “the Lubrizol patents.” I also understand that Baker Hughes requests that the United States Patent and Trademark Office (USPTO) cancel claims 1-11 of the 118 patent, claims 1-5 of the 498 patent, claims 1-5 of the 249 patent, and claims 1-9 of the 250 patent, i.e., all claims of the Lubrizol patents.

3. The claims of the 118 patent, the 498 patent, and the 249 patent are directed generally to a method of reducing the frictional pressure loss associated with the turbulent flow of a liquid hydrocarbon through a pipeline by introducing a drag reducing polymer into the liquid hydrocarbon. The claims of the 250 patent, on the other hand, are directed generally to a method of preparing a drag reducing

polymer that is capable of reducing the frictional pressure loss associated with the turbulent flow of a heavy crude oil through a pipeline. The claims of the Lubrizol patents specify certain properties of both the liquid hydrocarbon and the drag reducing polymer, which I will discuss below.

4. Throughout this declaration, I will refer to drag reducing polymers for use in liquid hydrocarbons as the relevant field or the relevant art.

5. In preparing this declaration, I have reviewed the Lubrizol patents and considered each of the documents cited in Section III in light of the general knowledge in the relevant art one year before the earliest filing date of the Lubrizol patents, i.e., one year before December 22, 2006. For ease of reference, every mention of “2005” means the time period in 2005 that is more than one year before the earliest filing date of the Lubrizol patents. In forming my opinions, I have relied upon my teaching, work, and research experience in polymer science and engineering, chemical engineering, and materials science.

## **II. Background and Qualifications**

6. A copy of my current (as of July 27, 2016) *curriculum vitae* is provided as Exhibit 1010. It provides a comprehensive description of my academic and employment history.

7. My education can be summarized as follows. I was awarded a B.S. degree in Chemical Engineering from the Massachusetts Institute of Technology

(MIT) in 1998, a M.S. degree in Chemical Engineering (Practice School) from MIT in 1999, and a Ph.D. in Chemical Engineering from the University of Minnesota, Twin Cities in 2004. I completed a National Research Council Postdoctoral Fellowship in the Polymer Physics Division at the National Institute of Standards and Technology (NIST) in 2006.

8. My academic employment record includes the following: From 2006 to 2012, I was an Assistant Professor at the University of Delaware (UD) in the Department of Chemical Engineering with a joint appointment in the Department of Materials Science & Engineering from 2011 to 2012. In 2012, I was promoted to Associate Professor with tenure at UD in the Department of Chemical & Biomolecular Engineering and Department of Materials Science & Engineering. I was awarded the chaired position of the Thomas and Kipp Gutshall Development Chair of Chemical & Biomolecular Engineering in 2012, and I retain that appointment currently. In 2016, I was promoted to Professor with tenure at UD in the Department of Chemical & Biomolecular Engineering and the Department of Materials Science & Engineering, which is my current position. My other academic appointments are listed in my *curriculum vitae*. Exhibit 1010.

9. Through my education and academic employment, I have gained significant expertise in the disciplines of polymer science, fluid dynamics, and thermodynamics. Because these three disciplines provide the basis for an

understanding of polymeric flow modifiers, including polymeric drag reducing agents, I have a specific knowledge and understanding regarding the operation of polymeric drag reducing additives to suppress the growth of turbulent eddies during fluid flow.

10. For example, four of the courses that I took as an undergraduate student at MIT were 10.000 (Chemical Engineering Seminar), 10.302 (Fluid Mechanics), 10.467 (Polymer Science Laboratory) and 10.691 (Polymer Physical Chemistry). The first two of the above courses were taught by Prof. Preetinder S. Virk, who is an expert in hydrocarbon flow in pipes, flow dynamics and streamlines, and drag reduction in fluids. In 10.000, the class had substantial discussion with respect to oil drilling, the challenges associated with an Alaska pipeline, the events that precipitated the Exxon Valdez incident, and the monetary and environmental repercussions of the Valdez incident. In 10.302, the class discussed laminar and turbulent flow properties, calculations of pressure drops, entrance effects and boundary layers in pipe flow, and modifications to flow profiles to minimize energy losses associated with fluid flow. In 10.467 (taught by Prof. Paula T. Hammond), the class synthesized polymers through free-radical and emulsion polymerization techniques, among many other activities. Additionally, the class measured the impact of the synthesized polymer additives on the flow properties of hydrocarbon fluids using an Ubbelohde viscometer. In 10.691

(taught by Prof. Ed W. Merrill, a pioneer in polymer synthesis), the class discussed methods for polymer synthesis, emulsion and suspension polymerization, radical polymerization, molecular weight and molecular weight distributions, methods to control and reduce molecular weight distributions, methods to generate high molecular weight polymers, polymer impacts on fluid flow properties (viscosity and intrinsic viscosity), and polymer solubility, among many other concepts. These courses provide a solid foundation in the concepts necessary to understand the behavior of drag reducing polymers in hydrocarbon fluids.

11. Additionally, several industrial experiences have allowed me to practice and refine my knowledge of polymer science, fluid dynamics, and thermodynamics, as relevant to polymer impacts on flow. For example, while consulting at General Electric (GE) Plastics, I optimized the flow of polymer mixtures into tools for injection molding with the goal of improving flow (reduced time and pressure) and ultimate part performance. During the subsequent time period, while consulting at Cargill, Inc., I worked on the design of a polylactide production facility. The activities associated with this assignment included obtaining pilot plant data on polymer reaction rates in solution and solution flow properties, modeling the data for scale-up, and generating a model of the production facility during startup and steady-state operation using the Aspen Dynamics and Aspen Plus software packages (including modeling and sizing

reactors, distillation columns, heat exchangers, pipes and pumps [and the pressure drops associated with fluid flow], and the ultimate energy input into the facility, etc.). Finally, I am currently engaged in activities with several companies in roles that take advantage of my understanding of polymer behavior in solution, including DuPont [government-funded industrial collaboration], Dow Chemical, Chemours, Synthomer (UK), and several other companies.

12. Furthermore, several experiences as a faculty member at the University of Delaware are relevant for understanding polymer effects on flow. One of the courses that I teach as a Professor at UD is cross-listed as CHEG [Chemical Engineering] 600 / MSEG [Material Science and Engineering] 630, Introduction to the Science and Engineering of Polymer Systems. In this course, one of the topics that I discuss is the synthesis of polymers through emulsion polymerization techniques, with an emphasis on methods to increase molecular weight and control polymer dispersity. Another topic that I address is polymer flow properties in solution and the effects of polymers on fluid flow (i.e. drag reduction, influence on shear thinning/thickening behavior, flow stability, etc.). Additionally, the course addresses polymer synthesis and polymer solubility as assessed through experimental and theoretical investigations of polymer solubility and solubility parameter calculations. Another course that I teach is CHEG 325, Chemical Engineering Thermodynamics II. In this course, one of the topics that I



address is solubility parameter calculations (through activity coefficient model development) and group contribution theory origins and methods.

13. Accordingly, I am an expert in the impact of polymer additives on the flow properties of fluids, and I have specific knowledge and understanding in the field of polymeric drag reducing additives.

14. Moreover, because the study of chemical engineering is closely tied to the petroleum industry, I have a general knowledge and understanding regarding the composition of crude oils. For example, most major chemical engineering separations courses, such as 10.37 (which I took while at MIT), have a strong foundation in separation processes associated with petroleum refining. In particular, distillation columns are a major focus, such that many institutions (including MIT and UD) have working, multi-story distillation columns housed within their buildings, on which students perform separation experiments with model compositions.

15. I am qualified to provide an opinion as to what a person of ordinary skill in the art would have understood, known, or concluded in 2005 because I was working in polymer science and engineering, chemical engineering, and materials science at that time and was familiar with the knowledge and education of others working in the field.

**III. Documents Considered in Forming My Opinions**

16. In forming my opinions, I have considered the following documents:

<b>Exhibit</b>	<b>Description</b>
1001	U.S. Patent No. 8,022,118 B2 (“the 118 patent”)
1002	U.S. Patent No. 8,426,498 B2 (“the 498 patent”)
1003	U.S. Patent No. 8,450,249 B2 (“the 249 patent”)
1004	U.S. Patent No. 8,450,250 B2 (“the 250 patent”)
1005	Holtmyer, Marlin D. and Chatterji, Jiten, Study of Oil Soluble Polymers as Drag Reducers, Polymer Engineering and Science, vol. 20, no. 7, 1980 (“the Holtmyer Publication”).
1006	U.S. Patent No. 3,758,406, Methods and Compositions for Reducing Frictional Pressure Loss in the Flow of Hydrocarbon Liquids, granted 9/11/1973 (“the Holtmyer Patent”)
1007	European Patent Application No. EP 0,882,739 A2, High molecular weight polymer and producing method the same and drag reducer, published 12/9/1998 (“Inaoka”)
1008	Carnahan, Norman F., “Precipitation of Asphaltenes in Heavy Oil and Tar Sands” in <i>Asphaltenes and Asphalts</i> , 2, Developments in Petroleum Science 40B, Elsevier Science B.V., 2000 (“Carnahan”)
1009	Strausz, O.P. and Lown, E.M., <i>The Chemistry of Alberta Oil Sands, Bitumens and Heavy Oils</i> , Alberta Energy Research Institute, Calgary, 2003 (“Strausz”)
1010	<i>Curriculum Vitae</i> of Thomas H. Epps III, Ph.D.
1011	Brandrup et al., <i>Polymer Handbook</i> , 4 <sup>th</sup> ed., John Wiley & Sons, Inc., 1999 (“Brandrup”)
1012	Young & Lovell, <i>Introduction to Polymers</i> , 2nd ed., CRC Press, 1991 (“Young & Lovell”)
1013	Odian, George, <i>Principles of Polymerization</i> , 4th ed., John Wiley & Sons, Inc., 2004 (“Odian”)
1014	Zakin, J.L. and Hunston, D.L., <i>Effect of Polymer Molecular Variables on Drag Reduction</i> , J. Macromol. Sci.-Phys., B18(4), 795-814 (1980)
1015	Mowla, D. and Naderi, A., <i>Experimental study of drag reduction by a polymeric additive in slug two-phase flow of crude oil and air in horizontal pipes</i> , Chemical Engineering

Exhibit	Description
	Science 61, 1549-1554, available online as of November 4, 2005
1016	Burger et al., <i>Studies of Drag Reduction Conducted over a Broad Range of Pipeline Conditions when Flowing Prudhoe Bay Crude Oil</i> , Journal of Rheology, 24(5), 603-626 (1980) (“Burger”)
1017	Buckley, J.S., et al., <i>Asphaltene Precipitation And Solvent Properties of Crude Oils</i> , Petroleum Science and Technology, vol. 16, no. 3-4, 1998 (“Buckley”)
1018	Ferguson, K.R., et al., Microbial Pilot Test for the Control of Paraffin and Asphaltenes at Prudhoe Bay, Society of Petroleum Engineers, Inc., 1996

#### IV. Person of Ordinary Skill in the Art

17. I understand that a person of ordinary skill in the art in 2005 is a hypothetical person who is presumed to be aware of all pertinent prior art as of that date, is familiar with the conventional wisdom in the art as of that date, and is a person of ordinary creativity.

18. Such a person would have been familiar with and routinely utilized basic principles relating to polymers and polymer synthesis, such as the chemical composition of monomers and polymers; common types of polymerization processes; and the properties of polymers, including solubility properties of polymers. He or she would regularly have consulted standard reference materials and texts in the field such as Brandrup et al., *Polymer Handbook*, 4th ed., John Wiley & Sons, Inc., 1999 (Ex. 1011; “Brandrup”); Young & Lovell, *Introduction*

to *Polymers*, 2nd ed., CRC Press, 1991 (Ex. 1012; “Young & Lovell”); Odian, George, *Principles of Polymerization*, 4th ed., John Wiley & Sons, Inc., 2004 (Ex. 1013; “Odian”).

19. Further, such a person would have been aware of and consulted technical and scientific publications specifically directed to the physical and chemical properties of drag reducing polymers and to the study of polymer flow properties in solution. Representative of such publications are Zakin, J.L. and Hunston, D.L., *Effect of Polymer Molecular Variables on Drag Reduction*, J. Macromol. Sci.-Phys., B18(4), 795-814 (1980) (Ex. 1014; “Zakin”); Mowla, D. and Naderi, A., *Experimental study of drag reduction by a polymeric additive in slug two-phase flow of crude oil and air in horizontal pipes*, Chemical Engineering Science 61, 1549-1554, available online as of November 4, 2005 (Ex. 1015; “Mowla”); and Burger et al., *Studies of Drag Reduction Conducted over a Broad Range of Pipeline Conditions when Flowing Prudhoe Bay Crude Oil*, Journal of Rheology, 24(5), 603-626 (1980) (Ex. 1016; “Burger”).

20. Such a person would have been readily able to determine properties of polymers, such as the weight average molecular weight of a polymer sample and the average number of repeat units in a polymer, using well-known methods. Such a person also would have been readily able to determine the solubility parameters of polymers, including using well-known approaches such as group contribution

theory calculations, enabling one to identify polymers that likely would be soluble in a target fluid and would therefore satisfy a major condition for use as drag reducing additives in the target fluid.

21. I have been asked to analyze the Lubrizol patents and the references discussed in this declaration from the perspective of a person of ordinary skill in the art as of 2005 – one year before the applications for the Lubrizol patents were filed.

22. In my opinion, in 2005 a person of ordinary skill in the art relating to the subject matter of the Lubrizol patents would typically have a B.S. degree in chemical engineering, polymer science and engineering, or a closely related field, with at least two years of work experience or three years of further academic experience with drag reducing polymers and/or the impact of polymer additives on the flow properties of fluids, and with a general knowledge of the composition and properties of liquid hydrocarbons such as crude oils.

23. My opinions set forth in this declaration are expressed from the perspective of a person of ordinary skill in the art as of 2005, as set forth above.

## **V. Drag Reducing Polymers**

24. The problem of friction-related drops in fluid pressure when transporting fluids, including for example crude oil, was well known to a person of ordinary skill in the art in 2005. Specifically, friction between a fluid and the walls

of a pipeline in which the fluid is flowing can result in the formation of turbulent eddies. Hydrocarbon fluids, such as crude oil, consist primarily of aromatic, naphthenic, and alkane-based molecules. The non-linear nature of the aromatics and naphthenics does not enable those molecules to appreciably dampen the turbulent eddies, which lead to significant pressure drops. These pressure drops correspond to a reduction in fluid flowrate through the pipeline.

25. As explained by the Lubrizol patents, the use of drag reducing polymers to reduce these friction-related drops in fluid pressure during the transportation of crude oil also was well understood by those of ordinary skill in the art in 2005. See, e.g., Ex. 1001 at 1:14-43; Ex. 1002 at 1:20-48; Ex. 1003 at 1:20-48; Ex. 1004 at 1:20-48.

26. Those of ordinary skill in the art in 2005 would have understood that drag reducing polymers lessen eddy-related drops in fluid pressure through a physical process. Drag reducing polymers typically are relatively long molecules with little branching. Therefore, in contrast with the non-linear structures of the components of the crude oil, the drag reducing polymers tend to stretch in the direction of fluid flow and absorb the energy of the turbulent eddies. By absorbing this energy, drag reducing polymers suppress the continued growth of the turbulent eddies.

27. Those of ordinary skill in the art in 2005 would have known that drag reducing polymers have very high molecular weights. Such drag reducing polymers typically were added to crude oil in extremely small quantities, *e.g.*, as parts per million by weight (referred to below as ppmw) of the crude oil. (All parts or parts per million expressed in this declaration are by weight unless otherwise indicated.) Because drag reducing polymers have very high molecular weights, they were known by those of ordinary skill in the art in 2005 to be effective at suppressing the growth of turbulent eddies even when present at such low concentrations as a few ppmw. See, for example, Ex. 1016 at 613.

28. Importantly, a person of ordinary skill in the art in 2005 would have understood that drag reducing polymers present as parts per million within the crude oil would have no appreciable effect on the overall composition and end-user properties of the crude oil. It would, on the other hand, have been relatively easy, for example, for one to simply dilute a crude oil with hexane to lessen friction-related drops in fluid pressure during the transportation of crude oil. Doing so, however, would alter the composition and ultimate end-user properties of the crude oil, and those of ordinary skill in the art in 2005 would have understood that such alteration may be commercially undesirable.

29. Also, it was generally understood in 2005 by persons of ordinary skill in the art that the effectiveness of a drag reducing polymer is related to its

solubility in the fluid being treated. The more soluble a drag reducing polymer is in the fluid, the greater the drag reducing effect of the polymer. See Ex. 1014 at 800-802. Indeed, in 2005, solubility in the fluid being treated had been referred to as the most important requirement of a drag reducing polymer. See Ex. 1015 at 1550.

30. Accordingly, for a polymer to effectively function as a drag reducing polymer in crude oil, a person of ordinary skill in the art in 2005 would have understood that the polymer should have (a) a straight chain structure with minimal branching, (b) high molecular weight, and (c) solubility in the crude oil into which it is to be incorporated.

## **VI. The Lubrizol Patents**

31. The Lubrizol patents, which are each entitled “Drag Reduction of Asphaltenic Crude Oils,” describe reducing the pressure drop associated with the formation of turbulent eddies in crude oil having a specified high asphaltene content and a specified low API gravity flowing through a pipeline. This reduction in pressure drop is said to be achieved by treating the crude oil with a drag reducing polymer. See Exs. 1001-1004.

32. As described by the Lubrizol patents, fluid in a pipeline must be transported with a sufficient fluid pressure to achieve the desired throughput. Drops in fluid pressure, however, are caused by turbulent eddies, such as those that



can result from friction between the wall of the pipeline and the fluid. The problems associated with these pressure drops are most acute when a fluid is transported over a long distance, such as through a pipeline. Ex. 1001 at 1:15-27; Ex. 1002 at 1:20-32; Ex. 1003 at 1:20-32; Ex. 1004 at 1:20-32.

33. As described by the Lubrizol patents, the use of high molecular weight polymeric drag reducing agents to substantially reduce the pressure drop was already well known to a person of ordinary skill in the art. More particularly, in 2005, a person of ordinary skill in the art would have known that high molecular weight polymeric drag reducing agents were widely used to suppress the growth of turbulent eddies, and that this suppression results in a higher flow rate of hydrocarbon liquid at a constant pumping pressure. Ex. 1001 at 1:28-39; Ex. 1002 at 1:33-44; Ex. 1003 at 1:33-44; Ex. 1004 at 1:33-44.

34. As also stated by the Lubrizol patents, it was known to a person of ordinary skill in the art that drag reduction “depends in part upon the molecular weight of the polymer additive and its ability to dissolve in the hydrocarbon under turbulent flow.” Ex. 1001 at 1:39-41; Ex. 1002 at 1:44-46; Ex. 1003 at 1:44-46; Ex. 1004 at 1:44-46. For example, as stated by the Lubrizol patents, effective drag reducing agents “typically have molecular weights in excess of five million.” Ex. 1001 at 1:41-43; Ex. 1002 at 1:46-48; Ex. 1003 at 1:46-48; Ex. 1004 at 1:46-48.

35. According to the Lubrizol patents, however, conventional polymeric drag reducing agents at that time “typically [did] not perform well in crude oils having a low API gravity and/or a high asphaltene content.” Ex. 1001 at 1:44-49; Ex. 1002 at 49-54; Ex. 1003 at 1:49-54; Ex. 1004 at 1:49-54.

36. As a result, the Lubrizol patents describe “a need for improved drag reducing agents capable of reducing the pressure drop associated with the turbulent flow of low API gravity and/or high-asphaltene crude oils through pipelines.” Ex. 1001 at 44-49; Ex. 1002 at 1:49-54; Ex. 1003 at 1:49-54; Ex. 1004 at 1:49-54. The claims of the Lubrizol patents reflect the goal of meeting this stated need. Specifically, each of the claims of the Lubrizol patents recites that the drag reducing polymer is (or is able to be) injected into a liquid hydrocarbon having an asphaltene content of at least 3 weight percent and an API gravity of less than about 26°. Exs. 1001, 1003, 1004 at claims; Ex. 1002 at claims (note, however, that some claims of the 498 patent merely recite that the liquid hydrocarbon have an asphaltene content of at least 3 weight percent or an API gravity of less than about 26°).

37. Asphaltenes are defined by the Lubrizol patents as “the fraction separated from crude oil or petroleum products upon addition of pentane . . . .” and, though difficult to characterize, are described generally as “high molecular

weight, non-crystalline, polar compounds which exist in crude oil.” Ex. 1001 at 2:64-3:8; Ex. 1002 at 2:54-65; Ex. 1003 at 2:54-65; Ex. 1004 at 3:5-17.

38. API gravity is defined by the Lubrizol patents as “the specific gravity scale developed by the American Petroleum Institute for measuring the relative density of various petroleum liquids.” Ex. 1001 at 3:59-4:6; Ex. 1002 at 3:49-63; Ex. 1003 at 3:49-63; Ex. 1004 at 4:1-15. Quite simply, a low API gravity represents a dense, or heavy, liquid. As a matter of common usage in 2005, the term “heavy crude” oil referred to oil having a low API gravity.

39. The API gravity recited in the claim merely indicates that the hydrocarbon liquid may be a heavy crude oil. As reflected by Table 1, reproduced below, oil having an API gravity of less than 26° is identified by the Lubrizol patents as heavy crude oil.

TABLE 1

Asphaltene Content, API Gravity, and Elasticity Response								
Crude Oil		ASPHALTENE		ELASTICITY RESPONSE				
				(AFFINITY)				
Sample	Type	CONTENT		API Gravity	LP	LP	Polymer A	Polymer B
		Test 1	Test 2		100	300		
Mercy	Heavy	16.8	15.5	16.0°	None	None	High	High
Petrozuata	Heavy	18.8	18.1	9.1°	None	None	High	High
Corocoro	Heavy	6.0	6.7	25.1°	None	None	High	High
Albian	Heavy	11.0	10.6	22.4°	None	None	High	High
Bow River	Heavy	11.4	10.3	21.8°	None	None	High	High
Maya	Heavy	14.6	15.4	21.9°	None	None	High	High
Western Canadian Select	Heavy	11.5	11.9	20.9°	None	None	High	High
San Joaquin Valley	Heavy	8.9	8.9	13.0°	None	None	High	High
Marlim Blend	Heavy	6.7	6.6	22.2°	High	High	High	High
West Texas Sour	Intermediate	2.8	2.8	31.6°	High	High	Moderate	Moderate
West Texas Intermediate	Light	0.5	—	41.6°	High	High	Moderate	Moderate
Basrah	Light	4.8	—	31.0°	High	High	Moderate	Moderate

40. The drag reducing polymer is defined in certain claims of the Lubrizol patents in terms of its molecular weight and its number of repeat units (which is related to molecular weight). Exs. 1001-1004 at claims. According to the Lubrizol patents, the drag reducing polymer has a weight average molecular weight of at least  $1 \times 10^6$  g/mol (grams per mole), i.e., a weight average molecular weight of at least one million g/mol. Ex. 1001 at claims 4, 10; Ex. 1002 at claims 4, 5; Ex. 1003 at claims 3, 5; Ex. 1004 at claims 4, 9.

41. The drag reducing polymer also is defined in the claims of the Lubrizol patents in terms of its solubility parameter. Exs. 1001, 1004 at claims. According to the Lubrizol patents, the drag reducing polymer has a solubility parameter that is similar to the solubility parameter of heavy crude oil. Compare, for example, Ex. 1001 at 4:33-36 (describing the liquid hydrocarbon as having a solubility parameter of at least about  $17 \text{ MPa}^{1/2}$ , or in the range of from about  $17.1 \text{ MPa}^{1/2}$  to about  $24 \text{ MPa}^{1/2}$ , or in the range of from  $17.5 \text{ MPa}^{1/2}$  to  $23 \text{ MPa}^{1/2}$ ) with Ex. 1001 at 12:24-28 (describing the drag reducing polymer as having a solubility parameter within identical ranges). The Lubrizol patents also state that the drag reducing polymer can have a solubility parameter that is within about  $4 \text{ MPa}^{1/2}$ , within about  $3 \text{ MPa}^{1/2}$ , or within  $2.5 \text{ MPa}^{1/2}$  of the solubility parameter of the liquid hydrocarbon, i.e., the heavy crude oil to be treated. Ex. 1001 at 12:24-31; Ex. 1002 at 12:1-12:8; Ex. 1003 at 11:65-12:5; Ex. 1004 at 12:26-33.

42. A person of ordinary skill in the art would have known in 2005 that the purpose of solubility parameters is to predict the solubility of one compound in another, or more generally, the potential of two compounds to mix, as opposed to separate into two chemically distinct phases. For example, it was generally understood in 2005 that the closer the solubility parameters of two materials, the more likely they are to form a homogenous solution. While a comparison of solubility parameters does not provide conclusive evidence of solubility, solubility

parameters were (and are) commonly used in this manner to predict solubility. Accordingly, by stating that a drag reducing polymer and a certain crude oil have similar solubility parameters, the Lubrizol patents would have simply indicated to a person of ordinary skill in the art in 2005 that the drag reducing polymer was thought to be soluble in the crude oil, which was already known to be important for achieving drag reduction as described above (see ¶29) and admitted in the background of the Lubrizol patents (Ex. 1001 at 1:39-41; Ex. 1002 at 1:44-46; Ex. 1003 at 1:44-46; Ex. 1004 at 1:44-46).

43. A person of ordinary skill in the art in 2005 also would have known and understood that there were various methods for determining solubility parameters, including, but not limited to, the methods set forth in the Lubrizol patents.

44. The Lubrizol patents disclose that the solubility parameter of the liquid hydrocarbon can be determined according to the equation  $\delta_2 = [(\Delta H_v - RT)/V]^{1/2}$  and the description found in Strausz, O. & Lown, M., *The Chemistry of Alberta Oil Sands, Bitumens and Heavy Oils* (Alberta Energy Research Institute), which was published in 2003. Ex. 1001 at 4:17-36; Ex. 1002 at 4:6-24; Ex. 1003 at 4:6-24; Ex. 1004 at 4:25-43. The method for determining a range of solubility parameters set forth in the Strausz publication referenced by the Lubrizol patents

would have been known by a person of ordinary skill in the art in 2005. See Ex. 1009 at 465-468; see also Ex. 1011 at 676 (identifying the above equation).

45. Similarly, the Lubrizol patents disclose that the solubility parameter of the drag reducing polymer is determined according to the group contribution method found in Brandrup et al., *Polymer Handbook* (4<sup>th</sup> ed., vol. 2, Wiley-Interscience), which was published in 1999. Ex. 1001 at 11:47-12:23; Ex. 1002 at 11:26-67; Ex. 1003 at 11:24-64; Ex. 1004 at 11:53-12:25. This group contribution method would have been known by a person of ordinary skill in the art in 2005. See Ex. 1011 at 677-686.

46. Additionally, a person of ordinary skill in the art in 2005 would have understood that a common route to increase the solubility parameter of a material is to introduce more heteroatom- (i.e. oxygen, sulfur, etc.) containing moieties into said material. As shown in Table 4 of Brandrup (See Ex. 1011 at 686), the addition of oxygen-containing groups such as a -COO-, a key constituent of [meth]acrylates, is expected to increase a material's overall solubility parameter ( $\delta$ ) to a much larger extent than the addition of non-polar groups such as -CH<sub>2</sub>-. A primary driver for this increase in  $\delta$  is the addition of a significant polar component ( $\delta_p$ ; i.e. increase in polarity) without a corresponding reduction in the dispersive component ( $\delta_d$ ). For example, a comparison of an ethyl group (-CH<sub>2</sub>-; a major component in polyalphaolefins such as polyethylene) vs. an ester group (-COO-; a

major component in methacrylates such as poly[2-ethylhexyl methacrylate]) shows that  $\delta_{d,-CH_2-}$  is  $270 \text{ J}^{1/2}\text{cm}^{3/2}/\text{mol}$  and  $\delta_{p,-CH_2-}$  is  $0 \text{ J}^{1/2}\text{cm}^{3/2}/\text{mol}$ , while  $\delta_{d,-COO-}$  is  $390 \text{ J}^{1/2}\text{cm}^{3/2}/\text{mol}$  and  $\delta_{p,-COO-}$  is  $490 \text{ J}^{1/2}\text{cm}^{3/2}/\text{mol}$ . The non-zero  $\delta_p$  for  $-COO-$  relative to  $-CH_2-$  is indicative of the increasing polar character of  $-COO-$  containing polymers, such as meth[acrylates].

47. Thus, a person of ordinary skill in the art would have expected that the overall solubility parameter of [meth]acrylate-based polymers would be increased relative to polyalphaolefins.

48. A person of ordinary skill in the art also would have understood that heavy crude oils typically have higher heteroatom contents than light or medium crude oils. This understanding is largely due to the higher amounts of asphaltene in heavy crude oil, asphaltene being known to contain nitrogen, oxygen, and sulfur heteroatoms. See for example Ex. 1009 at 464 (identifying asphaltene as the crude oil fraction having the highest “NOS” content, “NOS” standing for nitrogen, oxygen, sulfur). Accordingly, a person of ordinary skill in the art would have expected that heavy crude oil would have a higher solubility parameter than light or medium crude oil. Therefore, one would have expected that the introduction of oxygen heteroatoms to a drag reducing polymer likely would enhance the drag reducing polymer’s solubility in heavy crude oil (by increasing the drag reducing polymer’s solubility parameter to become closer to that of the heavy crude oil).



49. The Lubrizol patents describe the preparation and testing of two specific drag reducing [meth]acrylate-based polymers, identified as Polymer A and Polymer B. Polymer A is a poly(2-ethylhexyl methacrylate) homopolymer, which is described as having a solubility parameter of  $18.04 \text{ MPa}^{1/2}$ . Ex. 1001 at 13:12-36; Ex. 1002 at 12:55-13:11; Ex. 1003 at 12:52-13:10; Ex. 1004 at 13:12-36. Polymer B is a copolymer made from the copolymerization of 2-ethylhexyl methacrylate and *n*-butyl acrylate monomers, which is described as having a solubility parameter of  $20.55 \text{ MPa}^{1/2}$ . Ex. 1001 at 13:37-43; Ex. 1002 at 13:12-19; Ex. 1003 at 13:12-19; Ex. 1004 at 13:37-43.

50. The testing described by the Lubrizol patents involves a comparison of Polymer A and Polymer B to two commercially available polyalphaolefin drag reducing agents known as LP 100 and LP 300. LP 100 is described as having a solubility parameter of  $16.49 \text{ MPa}^{1/2}$  and LP 300 is described as having a solubility parameter of  $16.54 \text{ MPa}^{1/2}$ . Ex. 1001 at 13:46-14:11; Ex. 1002 at 13:23-37; Ex. 1003 at 13:23-37; Ex. 1004 at 13:46-14:11.

51. The Lubrizol patents describe the results of the comparative testing as follows:

[T]he results listed in Table 2 tend to show that the drag reduction achieved by addition of LP 100 product in light crude oil yields slightly more favorable results than either of the [experimental] products. However, when heavy crude oils are used, as shown in

Tables 3 and 4, the use of Polymers A or B results in higher percentages of drag reduction than either of the LP products.

Ex. 1001 at 18:18-24; Ex. 1002 at 17:50-56; Ex. 1003 at 17:45-51; Ex. 1004 at 18:12-18.

## **VII. The Holtmyer Publication**

52. The article titled *Study of Oil Soluble Polymers as Drag Reducers* was written by Marlin D. Holtmyer and Jiten Chatterji and published in the journal *Polymer Engineering and Science*, vol. 20, no. 7, in 1980 (Ex. 1005; “the Holtmyer Publication”). Holtmyer describes an investigation “to find the most effective material which would reduce the friction coefficient in turbulent flow when added in small quantities to oil pipelines.” Ex. 1005 at Abstract.

53. Because of its subject matter and its publication prior to 2005, a person of ordinary skill in the art considering the use of a drag reducing polymer in crude oil would have been familiar with the Holtmyer Publication.

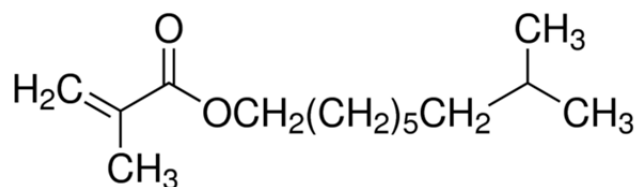
54. Holtmyer describes the problem of frictional pressure losses associated with the turbulent flow of a fluid through a conduit, stating “[w]hen a liquid is pumped under pressure, friction pressure is apparent as a pressure drop along the conduit. Such pressure drops are particularly noticeable under conditions where the velocity of liquid has surpassed the critical limit for laminar flow.” Ex. 1005 at p. 473, col. 1. Holtmyer explains that an appreciable reduction in friction

loss is desirable because “a decrease in friction loss would allow lower energy consumption or alternatively an increased flow rate under the original pumping conditions.” Ex. 1005 at p. 473, col. 1.

55. According to Holtmyer, a number of polymers had been used as drag reducing additives in crude oil, both in laboratory tests and large-scale field tests, but only one had received a certain degree of acceptance by 1980. Ex. 1005 at p. 473, col. 2. Accordingly, Holtmyer investigated the drag reduction properties of a series of homo- and copolymers of alkyl styrenes, acrylates, and methacrylates in hydrocarbon solvents to identify the most effective material for reducing frictional pressure losses associated with turbulent flow when added to oil pipelines. Ex. 1005 at p. 473, col. 2; abstract.

56. Among the polymers prepared, tested, and found by Holtmyer to be effective at reducing frictional pressure drop in hydrocarbon fluids is a poly(isodecyl methacrylate) homopolymer, referred to as “iDMA” by Holtmyer. Although it would be more technically accurate to abbreviate poly(isodecyl methacrylate) as PiDMA, I will refer to it throughout this declaration as iDMA to follow the convention of Holtmyer.

57. Isodecyl methacrylate – the monomeric constituent of the iDMA homopolymer – was known to contain oxygen heteroatoms. The chemical structure of the isodecyl methacrylate monomer is shown below:



Accordingly, a person of ordinary skill in the art would have understood that a plurality of the repeat units in the iDMA homopolymer contain oxygen heteroatoms. Indeed, because iDMA is a homopolymer that is made up solely of isodecyl methacrylate monomer segments, all of the repeat units of the iDMA homopolymer contain oxygen heteroatoms.

58. The polymers of Holtmyer, including the iDMA homopolymer, are prepared by emulsion polymerization. Ex. 1005 at 474, col. 1. In particular, the Holtmyer Publication describes a “standard procedure” in which a combination of water, sodium lauryl sulfate, potassium persulfate, sodium bisulfite, and freshly distilled monomer was agitated in a round bottom flask fitted with a mechanical stirrer, a thermometer, a gas inlet tube, and a Liebig condenser equipped with a gas outlet tube. The polymerization reaction was carried out in a nitrogen atmosphere for a period of 3 hours to 6 hours at 60 °C. The coagulated polymer was recovered by filtration and dried under vacuum. Ex. 1005 at p. 474, col. 1.

59. Emulsion polymerization is a polymerization technique in which the polymer is not soluble in the emulsion medium (usually water), whereas the initiator is only soluble in the emulsion medium. Typically, a surfactant is added

to the system, which upon agitation leads to stable monomer droplets that can polymerize to form into latex particles. Emulsion polymerization generally proceeds through 3 major steps: (1) latex particles are formed by monomer in the aqueous solution reacting with the initiator; (2) the number of particles remains constant and monomer diffuses from the medium into the polymerizing particles leading to an increase in molecular weight; (3) polymerization slows until all monomer is exhausted. See Ex. 1013 at 350-356.

60. A person of ordinary skill in the art would have understood that a major benefit of using emulsion polymerization to prepare drag reducing polymers is that it provides a facile method for the controlled production of high molecular weight polymers having a narrower distribution in individual chain molecular weights, also referred to as low dispersity. See Ex. 1013 at 351, 366.

61. As explained in the following paragraphs, a person of ordinary skill in the art would have understood the Holtmyer Publication to disclose an iDMA homopolymer having a weight average molecular weight that is well above the “at least  $1 \times 10^6$  g/mol” recited by certain claims of the Lubrizol patents.

62. In Table 6, Holtmyer describes a sample of iDMA having a viscosity average molecular weight of about  $26 \times 10^6$  g/mol, i.e. about 26 million g/mol. This sample is reported as performing exceptionally well with a minimum concentration effect, leading Holtmyer to state that this sample of iDMA “was

close to the optimum molecular weight for maximum performance.” Ex. 1005 at 475, col. 2.

63. For a low dispersity polymer such as those generated by Holtmyer using the emulsion polymerization technique described above, a person of ordinary skill in the art would have understood that the viscosity average molecular weight of an iDMA sample would be relatively close to, and not greater than, the weight average molecular weight of the sample. See, e.g., Ex. 1012 at 196-198. Accordingly, a person of ordinary skill in the art would have understood that the iDMA sample shown in Table 6 as having a viscosity average molecular weight of  $26 \times 10^6$  g/mol also would have a weight average molecular weight of at least  $26 \times 10^6$  g/mol (26 million g/mol), which is well above the “at least  $1 \times 10^6$  g/mol” recited by claims of the Lubrizol patents.

64. A person of ordinary skill in the art also would have understood that the iDMA sample identified by Holtmyer as having a viscosity average molecular weight of approximately 26 million g/mol in Table 6 would have contained well above the “at least about 25,000 repeating units” recited by certain claims of the Lubrizol patents.

65. The number of repeat units in a homopolymer can be calculated easily by dividing the number average molecular weight of the homopolymer by the molecular weight of a single repeat unit. A person of ordinary skill in the art

would easily have been able to identify (either through conventional resources or by simple calculation) the molecular weight of the isodecyl methacrylate repeat unit to be 226.36 g/mol.

66. A person of ordinary skill in the art would have understood that the number average molecular weight of a polymer sample could be calculated by dividing the weight average molecular weight of the polymer sample by the dispersity. For a low dispersity polymer such as that generated by Holtmyer using emulsion polymerization, a person of ordinary skill in the art would have understood that the number average molecular weight of an iDMA sample would be close to the weight average molecular weight of the sample. More particularly, a polymer such as that generated by Holtmyer using emulsion polymerization would have been understood to have a dispersity between about 1.1 and 2.0. See Ex. 1013 at 365-366 (identifying the upper boundary of the polydispersity index as 2.0). Therefore, a person of ordinary skill in the art would have understood that the number average molecular weight of the iDMA sample in Table 6 would be between at least about 13 million g/mol ( $26 \text{ million} / 2.0 \approx 13.0 \text{ million}$ ) and about 24 million g/mol ( $26 \text{ million} / 1.1 \approx 23.6 \text{ million}$ ).

67. By dividing the number average molecular weight of the iDMA homopolymer by the molecular weight of an isodecyl methacrylate repeat unit, a person of ordinary skill in the art would have easily calculated the number of

repeat units in the iDMA homopolymer to be at least between about 57,000 repeat units ( $13.0 \text{ million} / 226.36 \approx 57,400$  repeat units [dispersity of 2.0]) and about 104,000 repeat units ( $23.6 \text{ million} / 226.36 \approx 104,200$  repeat units [dispersity of 1.1]), which is well above the “at least about 25,000 repeating units” recited by certain claims of the Lubrizol patents.

68. As reported by Holtmyer, the iDMA homopolymer was subjected to a number of tests to determine its effectiveness as a drag reducing agent. Most of the testing was performed on a closed loop pipeline apparatus. Ex. 1005 at p. 474, col. 1 (stating that “[d]rag reduction properties of the polymers were evaluated by ... a small, closed-loop pipeline apparatus designed for one point pressure sensing.”). Such closed loop testing was (and is) generally understood by persons of ordinary skill in the art to be indicative of performance in a larger pipeline, such as a commercial oil pipeline.

69. For example, the drag reducing abilities of iDMA and polyisobutylene (the latter of which is identified by Holtmyer as an accepted drag reducing agent in crude oil) were compared in four different liquid hydrocarbons. In each instance, iDMA was found to be substantially more effective at achieving drag reduction. Ex. 1005 at p. 475, col. 2; Table 9.

70. As part of this testing, Holtmyer introduced iDMA into a QC-1156 solvent, described as “primarily an aromatic hydrocarbon with an API gravity of



22.5°”, and measured a 64% drag reduction. Ex. 1005 at Table 9; footnote to Table 9. Holtmyer also introduced iDMA into each of kerosene (64% drag reduction), Cardium crude oil (55% drag reduction), and Ellenberger crude oil (50% drag reduction). Table 9 shows that the introduction of iDMA into each of these liquid hydrocarbons significantly reduced the frictional pressure loss through the pipeline apparatus.

71. A person of ordinary skill in the art would have understood that the reduction in frictional pressure loss demonstrated by Table 9 is brought about by the ability of the iDMA drag reducing polymer to suppress the growth of turbulent eddies, that suppression being the mechanism by which drag reducing polymers were (and are) understood to function in turbulent flow by persons of ordinary skill in the art. See Ex. 1005 at abstract; see also Ex. 1001 at 1:28-37, Ex. 1002 at 1:33-42, Ex. 1003 at 1:33-42, Ex. 1004 at 1:33-42.

72. A person of ordinary skill in the art would have been led to add the iDMA drag reducing polymer to a liquid hydrocarbon at a concentration within the about 0.1 ppmw to about 500 ppmw range recited by certain claims of the Lubrizol patents. In particular, Holtmyer discloses that addition of the iDMA drag reducing polymer to a liquid hydrocarbon at a concentration of about 300 ppm provides a strong and efficient drag reduction effect.

73. Specifically, Table 6 in the Holtmyer Publication demonstrates that iDMA drag reducing polymers having each of two different molecular weights are effective when added to a liquid hydrocarbon (kerosene) at two concentrations: 600 ppm (0.06 wt.%) and 300 ppm (0.03 wt.%). Ex. 1005 at Table 6. Notably, for each of the two iDMA drag reducing polymers, introduction at 300 ppm resulted in significant drag reduction.

74. In fact, the drag reduction achieved by the iDMA polymer having a molecular weight of about 26 million g/mol (the higher molecular weight of the two iDMA polymers) was essentially the same when added at 300 ppm as when added at 600 ppm. Ex. 1005 at Table 6 (76% drag reduction at 300 ppm and 77% drag reduction at 600 ppm). This led Holtmyer to report that the higher molecular weight iDMA sample “performed exceptionally well with a minimum concentration effect.” Ex. 1005 at 475, col. 2.

75. The Holtmyer Publication also describes an evaluation of the concentration dependence of this “optimum molecular weight” sample of iDMA, with the results indicating that the drag reduction effect actually decreases as the concentration of the iDMA is increased from 300 ppm (0.03 wt.%) to 600 ppm (0.06 wt.%). Ex. 1005 at 475, col. 2; Table 8. Therefore, a person of ordinary skill in the art would have been led to add the high molecular weight iDMA polymer

disclosed by Holtmyer to a liquid hydrocarbon at a concentration of about 300 ppm to make efficient use of the material.

76. A person of ordinary skill in the art also would have understood that addition of the iDMA drag reducing polymer to a liquid hydrocarbon, such as the claimed liquid hydrocarbon, would not cause a decrease in the viscosity of the liquid hydrocarbon.

77. As a general matter, a person of ordinary skill in the art would have understood that the addition of high molecular weight polymers, such as those disclosed by Holtmyer would not cause a reduction in the viscosity of a liquid hydrocarbon. As shown by the Holtmyer Publication in Table 8, for example, adding increasing amounts of iDMA to a kerosene liquid hydrocarbon actually results in an *increase* in the viscosity of the treated liquid hydrocarbon. Ex. 1005 at Table 8. Accordingly, a person of ordinary skill in the art would have understood that addition of the high molecular weight iDMA drag reducing polymer of Holtmyer to a liquid hydrocarbon such as that defined by the claims of the Lubrizol patents would produce a treated liquid hydrocarbon in which the viscosity of the treated liquid hydrocarbon is not less than the viscosity of the liquid hydrocarbon prior to treatment with the drag reducing polymer.

78. As I noted above, certain claims (claims 1-2 and 4-5) of the 498 patent require simply that the liquid hydrocarbon have an asphaltene content of at least 3

weight percent **and/or** an API gravity of less than 26°. Each and every limitation of these claims is met by the testing described in Table 9 of the Holtmyer Publication.

79. Specifically, in Table 9 Holtmyer discloses the addition of an iDMA homopolymer to a QC-1156 solvent, described as being “primarily an aromatic hydrocarbon with an API gravity of 22.5°,” flowing through a “closed-loop pipeline apparatus.” Ex. 1005 at Table 9; p. 474, col. 1. Holtmyer also reports that the addition of the iDMA homopolymer to the QC-1156 liquid hydrocarbon resulted in a 64% reduction in drag. Ex. 1005 at Table 9. Accordingly, the results of the testing disclosed in Table 9 of the Holtmyer Publication meet each and every limitation of claim 1 of the 498 patent.

80. A person of ordinary skill in the art also would have understood that the iDMA homopolymer used in the testing disclosed in Table 9 of the Holtmyer Publication had a weight average molecular weight well above the “at least  $1 \times 10^6$  g/mol” recited by certain claims of the 498 patent.

81. The iDMA identified by Holtmyer in Table 9 is described as having an intrinsic viscosity of 5.5 dl/g at 23 °C in butyl acetate. Ex. 1005 at footnote to Table 9. According to Holtmyer, the relationship between intrinsic viscosity and the viscosity average molecular weight ( $M_v$ ) for the methacrylate polymers disclosed by Holtmyer can be approximated by the equation  $(\eta) = 8.64 \times 10^{-5}$

$M_v^{0.64}$ . Ex. 1005 at 474, col. 2. Although this equation represents the relationship between intrinsic viscosity and molecular weight for poly(lauryl methacrylate) in butyl acetate at 23 °C, Holtmyer suggests that this equation is generally valid for polymers of the methacrylate class, especially those listed in Table 1, under the same conditions of solvent and temperature. Ex. 1005 at 474, col. 2. Because the intrinsic viscosity of the iDMA homopolymer used in Table 9 is provided under the same conditions of solvent and temperature, a person of ordinary skill would have understood Holtmyer to teach that this equation is applicable to the iDMA homopolymer used in Table 9. Using this equation, I have calculated the viscosity average molecular weight of the iDMA homopolymer used in Table 9 to be approximately 32 million g/mol. My calculation appears in Attachment A.

82. As described above, for a low dispersity polymer such as those generated by Holtmyer, a person of ordinary skill in the art would have understood that the weight average molecular weight of an iDMA sample would be relatively close to, and not less than, the viscosity average molecular weight of the sample. See ¶60. Accordingly, a person of ordinary skill in the art would have understood that the iDMA sample used in Table 9 had a weight average molecular weight of at least 32 million g/mol, which is well above the “at least  $1 \times 10^6$  g/mol” recited by certain claims of the Lubrizol patents.

83. A person of ordinary skill in the art also would have understood that the iDMA sample used in the testing disclosed in Table 9 of the Holtmyer Publication contains well above the “at least about 25,000 repeating units” recited by certain claims of the 498 patent.

84. As described above, the number of repeat units in a homopolymer can be easily calculated by dividing the number average molecular weight of the homopolymer by the molecular weight of a single repeat unit, and a person of ordinary skill in the art would have been able to easily identify (either through conventional resources or by simple calculation) the molecular weight of the isodecyl methacrylate repeat unit to be 226.36 g/mol. See ¶62.

85. For low dispersity polymers such as those generated by Holtmyer, a person of ordinary skill in the art also would have understood that the number average molecular weight of an iDMA sample could be calculated by dividing the weight average molecular weight by the dispersity, which would be between about 1.1 and 2.0. See ¶63. Therefore, based on a weight average molecular weight of at least 32 million g/mol, a person of ordinary skill in the art would have understood that the number average molecular weight of the iDMA sample used in Table 9 would be between at least about 16 million g/mol ( $32 \text{ million} / 2.0 \approx 16.0 \text{ million}$ ) and about 29 million g/mol ( $32 \text{ million} / 1.1 \approx 29.1 \text{ million}$ ).

86. By dividing the number average molecular weight of the iDMA homopolymer by the molecular weight of an isodecyl methacrylate repeat unit, a person of ordinary skill in the art would have easily calculated the number of repeat units in the iDMA homopolymer to be between at least about 71,000 repeat units ( $16.0 \text{ million} / 226.36 \approx 70,700$  [dispersity of 2.0]) and about 128,000 repeat units ( $29.1 \text{ million} / 226.36 \approx 128,500$  [dispersity of 1.1]), which is well above the “at least about 25,000 repeating units” recited by certain claims of the 498 patent.

87. For these reasons, the testing disclosed in Table 9 of the Holtmyer Publication meets each and every element of claims 2, 4, and 5 of the 498 patent.

### **VIII. The Combination of the Holtmyer Publication and Carnahan**

88. The Holtmyer Publication does not explicitly disclose the introduction of the iDMA drag reducing polymer into a pipeline containing a flow of a crude oil having an asphaltene content of at least 3 weight percent and an API gravity of less than  $26^\circ$ , as recited by the claims of the Lubrizol patents. However, it would have been obvious to a person of ordinary skill in the art to have introduced the iDMA drag reducing polymer disclosed by Holtmyer into a pipeline containing a flow of crude oil having this combination of properties.

89. Crude oils containing an asphaltene content of at least 3 weight percent and an API gravity of less than  $26^\circ$  were well-known in 2005. See, for example, Ex. 1017 at 12, Table I (identifying Alaska-93, Alaska-95, Schuricht, and

Heavy California crude oils); Ex. 1018 at 557-558 (identifying Prudhoe Bay Alaskan crude oil); see also the Lubrizol patents, Exs. 1001-1004, at Table 1 (identifying a number of crude oils).

90. It also was well understood that there was (and is) a significant economic advantage to reducing the frictional pressure losses that occur during the transport of crude oils through pipelines. For instance, Holtmyer explained that a decrease in frictionally generally pressure loss would “allow lower energy consumption or alternatively an increased flow rate under the original pumping conditions,” rendering the use of drag reducing agents “economically profitable to industrial organizations engaged in movement of large volumes of liquid at high flow rates for considerable distances.” Ex. 1005 at p. 473, col. 1; see also Ex. 1001 at 1:15-30, Ex. 1002 at 1:20-35, Ex. 1003 at 1:20-35, Ex. 1004 at 1:20-35 (describing problems associated with frictional pressure drop and stating that drag reducing additives are commonly used in the industry to alleviate the problems).

91. A person of ordinary skill in the art would have recognized that these advantages would be realized in the transport of crude oil in general, which would include the “heavy” crude oil recited in the claims of the Lubrizol patents.

92. Accordingly, a person of ordinary skill in the art would have been motivated to introduce the iDMA drag reducing polymer disclosed by Holtmyer into a pipeline containing a flow of crude oil having the claimed asphaltene content



and API gravity properties to obtain the well-known economic benefits associated with drag reduction.

93. Moreover, a person of ordinary skill in the art would have had a reasonable expectation that the iDMA drag reducing polymer of Holtmyer would be effective at reducing the frictional pressure losses associated with turbulent flow when introduced into a crude oil having an asphaltene content of at least 3 weight percent and an API gravity of less than 26°, as recited by the claims of the Lubrizol patents.

94. In particular, Table 9 of the Holtmyer Publication demonstrates that the iDMA drag reducing polymer had a significant drag reducing effect in a liquid hydrocarbon having an API gravity less than about 26°. Specifically, the iDMA drag reducing polymer achieved a 64% drag reduction in QC-1156, which is disclosed as having an API gravity of 22.5°, well below the claimed 26° upper bound. Ex. 1005 at Table 9.

95. The Holtmyer Publication does not specifically disclose use of the iDMA in a hydrocarbon liquid containing at least 3 weight percent asphaltene (the asphaltene content of the QC-1156, Cardium crude oil, and Ellenberger crude oil in Table 9 is not disclosed). Accordingly, a person of ordinary skill in the art contemplating the use of the iDMA drag reducing polymer in a crude oil having an asphaltene content of at least 3 weight percent and an API gravity of less than 26°

would have been led to investigate the potential effect of asphaltene content on the ability of the iDMA polymer to achieve drag reduction. In particular, as described in detail below, a patent obtained by Holtmyer describing largely the same subject matter (including the iDMA drag reducing polymer) addresses the effect of asphaltene content on the ability of the iDMA polymer to achieve drag reduction.

96. U.S. Patent No. 3,758,406, titled “Methods and Compositions for Reducing Frictional Pressure Losses in the Flow of Hydrocarbon Liquids” and identifying William T. Malone, Marlin D. Holtmyer, John M. Tinsley, and Jiten Chatterji as inventors, issued in 1973 (Ex. 1006; “the Holtmyer Patent”). Because of its subject matter and publication prior to 2005, a person of ordinary skill in the art considering the use of a drag reducing polymer in crude oil (and particularly a person of ordinary skill in the art considering the use of a drag reducing polymer disclosed by the Holtmyer Publication) would have been familiar with the Holtmyer Patent.

97. In general, the Holtmyer Patent states that the optimum quantity of polymer to be introduced to a hydrocarbon liquid may vary depending on the type of liquid hydrocarbon involved, but that below a concentration of about .004 weight percent (40 ppm), insufficient polymer is present to effectively bring about a reduction in frictional pressure loss. Ex. 1006 at 4:51-59. The Holtmyer Patent also explains that when a solid agent is suspended in the liquid hydrocarbon, it is

preferable to use a somewhat larger amount of the polymer additive, such as from about 1 to about 10 pounds per 1000 gallons (between about 160 ppm and 1600 ppm). Ex. 1006 at 4:63-75.

98. Because asphaltenes were understood to be colloiddally dispersed (present as very small suspended particles) in crude oil, see for example Ex. 1008 at 326 and Ex. 1009 at 463, a person of ordinary skill in the art would have understood the Holtmyer Patent as teaching that crude oils having relatively high asphaltene contents may require the introduction of a higher amount of the drag reducing polymer than would be necessary for crude oils with low asphaltene contents.

99. Moreover, a person of ordinary skill in the art would have understood that if a particular drag reducing polymer has low solubility in a liquid hydrocarbon, it may not be possible to attain the relatively high concentrations described by the Holtmyer Patent (between about 160 ppm and about 1600 ppm). Therefore, to determine whether the iDMA drag reducing polymer could be added to a crude oil having an asphaltene content of at least 3 weight percent and an API gravity of less than 26° in the amounts described by the Holtmyer Patent as being effective for drag reduction, a person of ordinary skill in the art would have been led to evaluate the solubility of the iDMA drag reducing polymer in such a crude oil.

100. To evaluate the solubility of a drag reducing polymer in a particular liquid hydrocarbon such as a crude oil, a person of ordinary skill in the art would have understood that the solubility could either be tested by experimental methods or be predicted by comparing the solubility parameters of the two materials. Solubility parameters were (and are) commonly consulted to predict the solubility of one material in another without the need for time-consuming, and sometimes expensive, experimentation. Therefore, to easily evaluate the solubility of the iDMA drag reducing polymer in a crude oil having at least 3 weight percent asphaltene and an API gravity of less than about 26°, a person of ordinary skill in the art would have sought to compare the solubility parameters of the drag reducing polymer and the crude oil.

101. A person of ordinary skill in the art would have been able to easily identify the solubility parameter of the iDMA homopolymer of Holtmyer. For instance, the solubility parameters of many common polymers were published in well-known handbooks, such as the *Polymer Handbook* referenced by the Lubrizol patents (i.e., Brandrup). Ex. 1011 at 702-711; Table 10. Additionally, it was well understood in 2005 that one could readily calculate the solubility parameter of a polymer using the group contribution method presented in Brandrup. Ex. 1011 at 686-687; Table 4, 5.

102. I have calculated a value for the solubility parameter of the iDMA homopolymer of Holtmyer using the group contribution method presented in Brandrup at B11 and C30 (Ex. 1011 at 677, 683), i.e., the equations recited in the Lubrizol patents, as shown in Attachment B, and arrived at a solubility parameter value of about 17.84 MPa<sup>1/2</sup>.

103. One of the variables needed for calculating the solubility parameter of a polymer using the group contribution method of Brandrup is the molar volume of the polymer in question. Because neither the experimental molar volume nor the density (from which the molar volume could be calculated with knowledge of the compound's molecular weight) of the iDMA homopolymer is readily available from reliable sources, I calculated the molar volume of the iDMA homopolymer using the group contribution method described in Brandrup. Ex. 1011 at 682-683; Table 3. This calculation also is shown in Attachment B.

104. A person of ordinary skill in the art also would have been able to generally identify the solubility parameter of a crude oil having an asphaltene content of at least 3 weight percent and an API gravity of less than about 26°. A person of ordinary skill in the art would have recognized that, due to the complexity of crude oil, it would be very difficult to calculate the solubility parameter of such a crude oil using the group contribution methods described in Brandrup. Therefore, a person of ordinary skill in the art would have understood

that the solubility parameter of such a crude oil must be determined using other methods.

105. Because crude oil is a well-studied fluid, a person of ordinary skill in the art would have sought to locate the solubility parameters of crude oils having asphaltene contents of at least 3 weight percent and API gravities of less than about 26° within the published literature on the subject. A person of ordinary skill in the art would have expected that studies on the solubility (and solubility parameters) of crude oil (and crude oil components) had been performed because of the importance of maintaining the heavier components of crude oil, such as asphaltenes, dispersed/solubilized within the crude oil during transport (precipitation of those components within a conduit can severely restrict the flow of crude oil, which is highly undesirable).

106. The book titled *Asphaltenes and Asphalts*, 2, published in 2000 as part of an Elsevier Science B.V. series related to Developments in Petroleum Science, contains a chapter titled “Precipitation of Asphaltenes in Heavy Oil and Tar Sands” written by Norman F. Carnahan (Ex. 1008; “Carnahan”). Carnahan generally describes the study of asphaltene solubility in crude oils, including through solubility parameter models. Because of its subject matter and publication prior to 2005, a person of ordinary skill in the art would have looked to Carnahan as a potential source of solubility parameters for heavy, asphaltenic crude oils.

107. Indeed, having disclosed the solubility parameter of asphaltenes as being about 20 MPa<sup>1/2</sup> (Ex. 1008 at 324), Carnahan continues with a section titled “Solubility parameters of petroleum fluids.” Ex. 1008 at 325. Under this heading Carnahan states:

“Solubility parameter values for light crude components depend on composition, and are often in the range of about 6-8 hildebrands. . . . Solubility parameter values for medium and heavy oils are greater than those for light oils. Only a limited amount of reliable experimental data has been reported, although none for asphaltenes phase behavior in heavy oils and tar sands. The expected value of the solubility parameter of such heavy petroleum fluids is about 8-10 hildebrands.”

Ex. 1008 at 325. Hildebrand units ( $[\text{cal}/\text{cm}^3]^{1/2}$ ) are readily converted to MPa<sup>1/2</sup> units by multiplying by 2.046 (i.e.,  $1 [\text{cal}/\text{cm}^3]^{1/2} = 2.046 \text{ MPa}^{1/2}$ ). Accordingly, Carnahan teaches a person of ordinary skill in the art that the solubility parameters of heavy, asphaltenic crude oils are expected to fall within the range between about 16.4 MPa<sup>1/2</sup> and 20.5 MPa<sup>1/2</sup>.

108. Carnahan’s teaching is supported by the article titled *Asphaltene Precipitation And Solvent Properties of Crude Oils*, written by J.S. Buckley et al. and published in *Petroleum Science and Technology*, vol. 16, no. 3-4, in 1998 (Ex. 1017; “Buckley”). Buckley teaches that the solubility parameter of a crude oil can be estimated by using the refractive index of that crude oil. Because of its subject

matter and its publication prior to 2005, a person of ordinary skill in the art also would have looked to Buckley as a potential source of solubility parameters for asphaltenic crude oils.

109. Specifically, Buckley suggests that the solubility parameters of hydrocarbons have a linear relationship with a function of the refractive indexes of the hydrocarbons:  $F_{RI} = (n^2 - 1)/(n^2 + 2)$ . [Note that “n” is the standard abbreviation for refractive index; see also Ex. 1017 at 28 (identifying the symbol “n” as refractive index).] This relationship is shown graphically in Figure 1 of Buckley. Ex. 1017 at 4. Buckley also experimentally measured the refractive indexes (RI) of a number of heavy, asphaltenic crude oils. Ex. 1017 at 12 (Table I). By substituting the RI values for those oils into the above equation and plotting the results in Figure 1, I was able to estimate the solubility parameters of the following crude oils using the teachings of Buckley:

- A-93 crude oil (API gravity 25.5°; n-C<sub>5</sub> asphaltenes 10.9%):

$$RI = 1.522; F_{RI} \approx 0.305; \text{solubility parameter} \approx 18 \text{ MPa}^{1/2}$$

- A-95 crude oil (API gravity 25.2°; n-C<sub>5</sub> asphaltenes 5.0%):

$$RI = 1.518; F_{RI} \approx 0.303; \text{solubility parameter} \approx 18 \text{ MPa}^{1/2}$$

- Schuricht crude oil (API gravity 24.6°; n-C<sub>5</sub> asphaltenes 9.9%):

$$RI = 1.514; F_{RI} \approx 0.301; \text{solubility parameter} \approx 18 \text{ MPa}^{1/2}$$

- Heavy CA crude oil (API gravity 19.0°; n-C<sub>5</sub> asphaltenes 6.8%):



$$RI = 1.565; F_{RI} \approx 0.326; \text{solubility parameter} \approx 20 \text{ MPa}^{1/2}$$

Accordingly, a person of ordinary skill in the art would have understood from Buckley that the solubility parameters of crude oils having an asphaltene content of at least 3 weight percent and an API gravity less than  $26^\circ$  were expected to fall squarely within the range taught by Carnahan.

110. Because the solubility parameter of the iDMA homopolymer described by Holtmyer ( $17.84 \text{ MPa}^{1/2}$ ) falls squarely within the range taught by Carnahan (about  $16.4 \text{ MPa}^{1/2}$  to  $20.5 \text{ MPa}^{1/2}$ ), a person of ordinary skill in the art would have had a reasonable expectation that the iDMA polymer would be soluble in a heavy crude oil having an asphaltene content of at least 3 weight percent. Therefore, a person of ordinary skill in the art would have expected that the iDMA homopolymer could be introduced into such a crude oil at the concentrations disclosed by the Holtmyer Patent as being effective to reduce frictional pressure loss in a liquid hydrocarbon containing suspended solids.

111. Because iDMA (a) was shown by Holtmyer to achieve significant drag reduction in a liquid hydrocarbon having an API gravity less than  $26^\circ$  and (b) would have been expected to be soluble in a liquid hydrocarbon having an asphaltene content of at least 3 weight percent at the concentrations disclosed by the Holtmyer Patent as being effective, a person of ordinary skill in the art would have had a reasonable expectation that the iDMA drag reducing polymer would be

effective at achieving drag reduction in a liquid hydrocarbon having an API gravity less than about 26° and an asphaltene content of at least 3 weight percent undergoing turbulent flow through a pipeline.

#### **IX. The Combination of the Holtmyer Publication and Strausz**

112. The Holtmyer Publication does not explicitly disclose the introduction of the iDMA drag reducing polymer into a pipeline containing a flow of a crude oil having an asphaltene content of at least 3 weight percent and an API gravity of less than 26°, as recited by the claims of the Lubrizol patents. However, it would have been obvious to a person of ordinary skill in the art to have introduced the iDMA drag reducing polymer disclosed by Holtmyer into a pipeline containing a flow of crude oil having the recited combination of properties.

113. Crude oils containing an asphaltene content of at least 3 weight percent and an API gravity of less than 26° were well-known in 2005. See, for example, Ex. 1017 at 12, Table I (identifying Alaska-93, Alaska-95, Schuricht, and Heavy California crude oils); Ex. 1018 at 557-558 (identifying Prudhoe Bay Alaskan crude oil); see also the Lubrizol patents, Exs. 1001-1004, at Table 1 (identifying a number of crude oils).

114. It also was well understood that there was (and is) a significant economic advantage to reducing the frictional pressure losses that occur during the transport of crude oils through pipelines. For instance, Holtmyer explained that a

decrease in frictionally generally pressure loss would “allow lower energy consumption or alternatively an increased flow rate under the original pumping conditions,” rendering the use of drag reducing agents “economically profitable to industrial organizations engaged in movement of large volumes of liquid at high flow rates for considerable distances.” Ex. 1005 at p. 473, col. 1; see also Ex. 1001 at 1:15-30, Ex. 1002 at 1:20-35, Ex. 1003 at 1:20-35, Ex. 1004 at 1:20-35 (describing problems associated with frictional pressure drop and stating that drag reducing additives are commonly used in the industry to alleviate the problems).

115. A person of ordinary skill in the art would have recognized that these advantages would be realized in the transport of crude oil in general, which would include the “heavy” crude oil recited in the claims of the Lubrizol patents.

116. Accordingly, a person of ordinary skill in the art would have been motivated to introduce the iDMA drag reducing polymer disclosed by the Holtmyer Publication into a pipeline containing a flow of crude oil having the claimed asphaltene content and API gravity properties to obtain the well-known economic benefits associated with drag reduction.

117. Moreover, a person of ordinary skill in the art would have had a reasonable expectation that the iDMA drag reducing polymer of Holtmyer would be effective at reducing the frictional pressure losses associated with turbulent flow when introduced into a crude oil having an asphaltene content of at least 3 weight

percent and an API gravity of less than 26°, as recited by the claims of the Lubrizol patents.

118. In particular, Table 9 of the Holtmyer Publication demonstrates that the iDMA drag reducing polymer had a significant drag reducing effect in a liquid hydrocarbon having an API gravity of less than about 26°. Specifically, the iDMA drag reducing polymer achieved a 64% drag reduction in QC-1156, which is disclosed as having an API gravity of 22.5°, well below the claimed 26° upper bound. Ex. 1005 at Table 9.

119. The Holtmyer Publication does not specifically disclose use of the iDMA in a hydrocarbon liquid containing at least 3 weight percent asphaltene (the asphaltene content of the QC-1156, Cardium crude oil, and Ellenberger crude oil in Table 9 is not disclosed). Accordingly, a person of ordinary skill in the art contemplating the use of the iDMA drag reducing polymer in a crude oil having an asphaltene content of at least 3 weight percent and an API gravity of less than 26° would have been led to investigate the potential effect of asphaltene content on the ability of the iDMA polymer to achieve drag reduction. In particular, as described in detail below, a patent obtained by Holtmyer describing largely the same subject matter (including the iDMA drag reducing polymer) addresses the effect of asphaltene content on the ability of the iDMA polymer to achieve drag reduction.

120. U.S. Patent No. 3,758,406, titled “Methods and Compositions for Reducing Frictional Pressure Losses in the Flow of Hydrocarbon Liquids” and identifying William T. Malone, Marlin D. Holtmyer, John M. Tinsley, and Jiten Chatterji as inventors, issued in 1973 (Ex. 1006; “the Holtmyer Patent”). Because of its subject matter and its publication prior to 2005, a person of ordinary skill in the art considering the use of a drag reducing polymer in crude oil (and particularly a person of ordinary skill in the art considering the use of a drag reducing polymer disclosed by the Holtmyer Publication) would have been familiar with the Holtmyer Patent.

121. In general, the Holtmyer Patent states that the optimum quantity of polymer to be introduced to a hydrocarbon liquid may vary depending on the type of liquid hydrocarbon involved, but that below a concentration of about .004 weight percent (40 ppm), insufficient polymer is present to effectively bring about a reduction in frictional pressure loss. Ex. 1006 at 4:51-59. The Holtmyer Patent also explains that when a solid agent is suspended in the liquid hydrocarbon, it is preferable to use a somewhat larger amount of the polymer additive, such as from about 1 to about 10 pounds per 1000 gallons (between about 160 ppm and 1600 ppm). Ex. 1006 at 4:63-75.

122. Because asphaltenes were understood to be colloiddally dispersed (present as very small suspended particles) in crude oil, see for example Ex. 1008

at 326 and Ex. 1009 at 463, a person of ordinary skill in the art would have understood the Holtmyer Patent as teaching that crude oils having relatively high asphaltene contents may require the introduction of a higher amount of the drag reducing polymer than would be necessary for crude oils with low asphaltene contents.

123. Moreover, a person of ordinary skill in the art would have understood that if a particular drag reducing polymer has low solubility in a liquid hydrocarbon, it may not be possible to attain the relatively high concentrations described by the Holtmyer Patent as being effective (between about 160 ppm and about 1600 ppm). Therefore, to determine whether the iDMA drag reducing polymer could be added to a crude oil having an asphaltene content of at least 3 weight percent and an API gravity of less than 26° in the amounts described by the Holtmyer Patent as being effective for drag reduction, a person of ordinary skill in the art would have been led to evaluate the solubility of the iDMA drag reducing polymer in such a crude oil.

124. To evaluate the solubility of a drag reducing polymer in a particular liquid hydrocarbon such as a crude oil, a person of ordinary skill in the art would have understood that the solubility could either be tested by experimental methods or be predicted by comparing the solubility parameters of the two materials. Solubility parameters were (and are) commonly consulted in order to predict the

solubility of one material in another without the need for time-consuming, and sometimes expensive, experimentation. Therefore, to easily evaluate the solubility of the iDMA drag reducing polymer in a crude oil having at least 3 weight percent asphaltene and an API gravity of less than about 26°, a person of ordinary skill in the art would have sought to compare the solubility parameters of the drag reducing polymer and the crude oil.

125. A person of ordinary skill in the art would have been able to easily identify the solubility parameter of the iDMA homopolymer of Holtmyer. For instance, the solubility parameters of many common polymers were published in well-known handbooks, such as the *Polymer Handbook* referenced by the Lubrizol patents (i.e., Brandrup). Ex. 1011 at 702-711; Table 10. Additionally, it was well understood in 2005 that one could readily calculate the solubility parameter of a polymer using the group contribution method presented in Brandrup. Ex. 1011 at 686-687; Table 4, 5.

126. I have calculated a value for the solubility parameter of the iDMA homopolymer of Holtmyer using the group contribution method presented in Brandrup at B11 and C30 (Ex. 1011 at 677, 683), i.e., the equations recited in the Lubrizol patents, as shown in Attachment B, and arrived at a solubility parameter value of about 17.84 MPa<sup>1/2</sup>.

127. One of the variables needed for calculating the solubility parameter of a polymer using the group contribution method of Brandrup is the molar volume of the polymer in question. Because neither the experimental molar volume nor the density (from which the molar volume could be calculated with knowledge of the compound's molecular weight) of the iDMA homopolymer is readily available from reliable sources, I calculated the molar volume of the iDMA homopolymer using the group contribution method described in Brandrup. Ex. 1011 at 682-683; Table 3. This calculation also is shown in Attachment B.

128. A person of ordinary skill in the art also would have been able to generally determine a range of solubility parameters for a crude oil having an asphaltene content of at least 3 weight percent. A person of ordinary skill in the art would have recognized that, due to the complexity of crude oil, it would be very difficult to calculate the solubility parameter of such a crude oil using the group contribution methods described in Brandrup. Therefore, a person of ordinary skill in the art would have understood that the solubility parameter of such a crude oil must be determined using other methods.

129. Crude oil is a complex fluid containing a number of different components. When faced with determining the solubility parameter of such a fluid, a person of ordinary skill in the art would have looked to solubility



characteristics of its individual components to determine a range within which the solubility parameter of the crude oil falls.

130. In particular, a person of ordinary skill in the art would start by looking at the solubilized and dispersed components of crude oil. The solubilized and dispersed constituents of crude oil include resins, asphaltenes, and cokes. By determining the range of fluids (that have known or easily calculated solubility parameters) that are capable of solubilizing or dispersing these crude oil components, a person of ordinary skill in the art would obtain a range of solubility parameters within which a heavy crude oil—in which these constituents are solubilized or dispersed in relatively high amounts compared to a light crude oil—would fall.

131. To obtain a (relatively) narrow range within which a solubility parameter of a heavy crude oil falls, a person of ordinary skill in the art would have looked toward the solubility characteristics of the higher molecular weight components of crude oil, such as asphaltene. The rationale is as follows: as the molecular weight of the soluble (dispersed) components increases, the range of solubility parameters for fluids capable of solubilizing (dispersing) these higher molecular weight components narrows. Therefore, looking to the solubility characteristics of the higher molecular weight components, such as asphaltene, would allow a person of ordinary skill in the art to refine significantly the range for

the solubility parameter of a heavy crude oil. Note that one would have been deterred from performing this analysis on the highest molecular weight constituents of crude oil, i.e. cokes, because they would be very difficult to solubilize (disperse) in easily studied fluids.

132. A person of ordinary skill in the art also would have been familiar with this type of experimentation. This type of experimentation is (was) often referred to as solvent fractionation. Among other things, it was used to refine the solubility parameter value of a complex or unknown material.

133. The book titled *The Chemistry of Alberta Oil Sands, Bitumens and Heavy Oils*, was written by Otto P. Strausz and Elizabeth M. Lown, and published by Alberta Energy Research Institute in 2003 (Ex. 1009; “Strausz”). Strausz addresses a number of topics related to the composition and properties of crude oils. In Chapter 14, Strausz describes the properties of asphaltene, including its solubility characteristics. Ex. 1009 at 464. Because of its subject matter and its publication prior to 2005, a person of ordinary skill in the art looking to determine the solubility parameter of a heavy, asphaltenic crude oil would have been led to Strausz.

134. Strausz generally describes the relationship between the molecular weight of the components of crude oil and the range of fluids that are capable of solubilizing each component. Specifically, Strausz states “solvents for coke

represent a subset of solvents for asphaltene, asphaltene for resins, resins for aromatics meaning that solvents for asphaltene are solvents for resins, etc....” Ex. 1009 at 473. This relationship also is shown graphically by Strausz. See Ex. 1009 at 474 (note the narrowing range of fluids in which the component is soluble as one moves from the lower molecular weight aromatics to the intermediate molecular weight resins and on to the higher molecular weight asphaltenes).

135. Although Strausz states that asphaltene forms a colloidal system in crude oil, and would therefore technically be “dispersed” rather than “soluble” in crude oil, Strausz recognizes that the solubility parameter model is nevertheless applicable in such a system. Accordingly, Strausz generally describes the ability of a liquid, such as crude oil, to disperse asphaltene in terms of “solubility”. See Ex. 1009 at 464 (“The solubility of asphaltene in the maltene of crude oil and the stability of the asphaltene solution with respect to precipitation are of considerable interest to all phases of the petroleum industry....”). For consistency, I have used the same terminology as used by Strausz.

136. Strausz describes the solubility characteristics of the high molecular weight asphaltenes in terms of solubility parameter. In particular, Strausz describes a study of the correlation between the solubility of asphaltene in a variety of test solvents and the solubility parameter of each of those test solvents. Ex. 1009 at 467-468. The study uses the equation  $\delta_2 = [(\Delta H_v - RT)/V]^{1/2}$  for calculating

the solubility parameters of the test solvents. Ex. 1009 at 468, Table 14.1. Based on that study, Strausz states that “the solvation energy of the solvents with  $\delta_2$  in the 17.1-22.1 MPa<sup>1/2</sup> range is sufficiently large to overcome the cohesion energy of asphaltene and cause solubilization.” Ex. 1009 at 467. Therefore, Strausz teaches that to solubilize the typical asphaltene contents of heavy crude oils, a hydrocarbon fluid should have a solubility parameter between about 17.1 MPa<sup>1/2</sup> and about 22.1 MPa<sup>1/2</sup> (as determined using the above equation).

137. A person of ordinary skill in the art would have understood from Strausz that, generally, for a typical heavy crude oil flowing through a pipeline to have had an asphaltene content of at least 3 weight percent, that crude oil would have a solubility parameter within the disclosed range of about 17.1 MPa<sup>1/2</sup> to about 22.1 MPa<sup>1/2</sup>.

138. Because the solubility parameter of the iDMA homopolymer described by Holtmyer (17.84 MPa<sup>1/2</sup>) falls squarely within the range taught by Strausz (17.1 MPa<sup>1/2</sup> and 22.1 MPa<sup>1/2</sup>), a person of ordinary skill in the art would have had a reasonable expectation that the iDMA homopolymer would be soluble in a crude oil having an asphaltene content of at least 3 weight percent. Therefore, a person of ordinary skill in the art would have expected that the iDMA homopolymer could be introduced into such a crude oil at the concentrations

disclosed by the Holtmyer Patent as being effective to bring about a reduction in frictional pressure loss in a liquid hydrocarbon containing suspended solids.

139. Because iDMA (a) was shown by the Holtmyer Publication to achieve significant drag reduction in a liquid hydrocarbon having an API gravity of less than 26° and (b) would have been predicted to be soluble in a liquid hydrocarbon having an asphaltene content of at least 3 weight percent at the concentrations disclosed by the Holtmyer Patent as being effective, a person of ordinary skill in the art would have had a reasonable expectation that the iDMA drag reducing polymer would be effective at achieving drag reduction in a liquid hydrocarbon having both an API gravity of less than about 26° and an asphaltene content of at least 3 weight percent undergoing turbulent flow through a pipeline.

140. Further, based on the teaching of Strausz, including the table providing solubility parameters for many of the components of crude oil (Ex. 1009 at 468, Table 14.1), a person of ordinary skill in the art would have understood that at least some, and likely a fairly large proportion, of the crude oils having an API gravity of less than about 26° and an asphaltene content of at least 3 weight percent would have solubility parameters between about 17.1 MPa<sup>1/2</sup> (the lower bound of the range disclosed by Strausz, which is within 1 MPa<sup>1/2</sup> of the solubility parameter of the iDMA homopolymer) and 21.84 MPa<sup>1/2</sup> (which is within 4 MPa<sup>1/2</sup> of the solubility parameter of the iDMA homopolymer). A person of ordinary skill in the

art would therefore have understood that at least some, and likely a fairly large proportion, of these crude oils would have solubility parameters within  $4 \text{ MPa}^{1/2}$  of the solubility parameter of the iDMA homopolymer.

141. Similarly, a person of ordinary skill in the art would have understood that at least some, and likely a fairly large proportion, of the crude oils having an API gravity of less than about  $26^\circ$  and an asphaltene content of at least 3 weight percent would have a solubility parameter between  $17.1 \text{ MPa}^{1/2}$  (the lower bound of the range disclosed by Strausz, which is within  $1 \text{ MPa}^{1/2}$  of the solubility parameter of the iDMA homopolymer) and  $20.34 \text{ MPa}^{1/2}$  (which is within  $2.5 \text{ MPa}^{1/2}$  of the solubility parameter of the iDMA homopolymer). A person of ordinary skill in the art would therefore have understood that at least some, and likely a fairly large proportion, of these crude oils would have solubility parameters within  $2.5 \text{ MPa}^{1/2}$  of the solubility parameter of the iDMA homopolymer.

142. In general, it would have been obvious in 2005 for a person of ordinary skill in the art to select a drag reducing polymer for a crude oil in which the drag reducing polymer was expected to be most effective, i.e., to provide the highest degree of drag reduction. It was well known in the art that the closer the solubility parameter of a drag reducing polymer is to that of a liquid hydrocarbon, the more soluble that one would have expected the polymer to be in that hydrocarbon, and thus the higher the degree of drag reduction that would have

been expected. To achieve the greatest drag reduction, therefore, it would have been obvious for one of ordinary skill in the art to have selected a drag reducing polymer for use in a crude oil in which it was expected to be highly soluble, i.e., a crude oil having a close solubility parameter. Indeed, selecting a drag reducing polymer that would produce the greatest drag reducing effect would have been a matter of routine optimization.

143. Accordingly, it would have been obvious for a person of ordinary skill in the art to have utilized the iDMA of Holtmyer in a crude oil having a solubility parameter within  $4 \text{ MPa}^{1/2}$ , and more preferably within  $2.5 \text{ MPa}^{1/2}$ , of the solubility parameter of the iDMA drag reducing polymer.

144. By reciting that the solubility parameters of the drag reducing agent and the liquid hydrocarbon are close to one another, the Lubrizol patents do no more than use solubility parameters (in a conventional manner) to describe the already well-known relationship between the solubility of a drag reducing polymer (such as those discussed herein) in a fluid and the expected effectiveness of the drag reducing polymer in that fluid. In fact, Lubrizol's selection of  $4 \text{ MPa}^{1/2}$  as the claimed range corresponds with Strausz's description that, typically, materials whose solubility parameters differ by greater than  $4 \text{ MPa}^{1/2}$  are largely immiscible or insoluble. Ex. 1009 at 466 ("liquids differing by two hildebrands [i.e., about 4

MPa<sup>1/2</sup>] in their [solubility parameter] values are generally incompletely miscible....”).

## **X. The Inaoka Patent Application Publication**

145. European Patent Application No. EP 0,882,739 A2, titled “High molecular weight polymer and producing method the same and drag reducer” and identifying Toru Inaoka et al. as inventors, was published in 1998 (Ex. 1007; “Inaoka”). Inaoka describes a polymer that is suitably adopted as a drag reducer and that has (a) a higher molecular weight than a conventional polymer, (b) a straight-chain structure with less branching than a conventional polymer, and (c) solubility in an organic solvent. Ex. 1007 at Abstract.

146. Because of its subject matter and publication prior to 2005, a person of ordinary skill in the art considering the use of a drag reducing polymer in crude oil would have been familiar with Inaoka.

147. Inaoka describes the use of polymeric drag reducers in crude oil flowing through a pipeline. Specifically, Inaoka states, “when an organic liquid such as crude oil is transported through a conduit such as a pipeline, there is a case that the transport of the liquid is not carried out smoothly.” Ex. 1007 at p. 3, lines 9-10. Inaoka also explains that this issue is especially problematic in long distance transport because transportation pressure is lost in a short distance, requiring a large number of pumping stations. Ex. 1007 at p. 3, lines 10-13.



148. To suppress this loss of transportation pressure, Inaoka describes the conventional use of a drag reducer that includes a high molecular weight polymer. Ex. 1007 at p. 3, lines 14-17. According to Inaoka, it is known that the polymer should have a molecular weight as high as possible, with molecular weights of 1.8 million g/mol to 7.4 million g/mol being known to exhibit drag reduction. Ex. 1007 at p. 3, lines 18-21. Additionally, the polymer should have a straight-chain structure with minimal branching. Ex. 1007 at p. 3, lines 18-19.

149. Inaoka describes that such polymers are known to be produced by the method disclosed in U.S. Patent No. 3,758,406. Ex. 1007 at p. 3, lines 22-31. U.S. Patent No. 3,758,406, titled “Methods and Compositions for Reducing Frictional Pressure Losses in the Flow of Hydrocarbon Liquids” and identifying William T. Malone, Marlin D. Holtmyer, John M. Tinsley, and Jiten Chatterji as inventors, issued in 1973 (Ex. 1006; “the Holtmyer Patent”).

150. The Holtmyer Patent largely describes the same subject matter and testing disclosed in the Holtmyer Publication, which I have discussed above in Section VII. In particular, it “relates to methods and compositions for reducing the frictional pressure loss encountered in the turbulent flow of hydrocarbon liquids through a conduit.” Ex. 1006 at Abstract. More specifically, the Holtmyer Patent describes the preparation and use of a polymeric drag reducing agent that is “a homopolymer or a copolymer of alkyl acrylate and methacrylate monomers having

ester groups containing from 4 to 18 carbon atoms or copolymers of such monomers and certain styrene, acrylate or methacrylate type monomers.” Ex. 1006 at Abstract.

151. According to Inaoka, many of the polymeric drag reducing agents prepared by the method disclosed in the Holtmyer Patent are difficult to produce with sufficiently high molecular weight. Ex. 1007 at p. 3, lines 27-31. Inaoka states that this has given rise to a need for a method for efficiently producing a high molecular weight polymer that is “suitably adopted as a drag reducer”. Ex. 1007 at p. 3, lines 27-31. According to Inaoka, his disclosure provides this method. Ex. 1007 at p. 3, lines 35-51. Thus, a person of ordinary skill in the art would have read Inaoka as attempting to improve upon the polymeric drag reducing agents disclosed by the Holtmyer Patent.

152. Inaoka teaches that “since the high molecular weight polymer in accordance with the present invention has a higher molecular weight than the conventional polymer, and has a straight-chain structure with less branching, and is soluble in an organic solvent, it is possible to provide a drag reducer having higher quality than the conventional drag reducer.” Ex. 1007 at p. 4, lines 4-7; see also Ex. 1007 at p. 17, lines 1-5. Accordingly, Inaoka explains, “[t]he drag reducer can be suitably adopted in transporting of an organic liquid such as crude oil through a conduit, such as a pipeline.” Ex. 1007 at p. 17, lines 1-5.

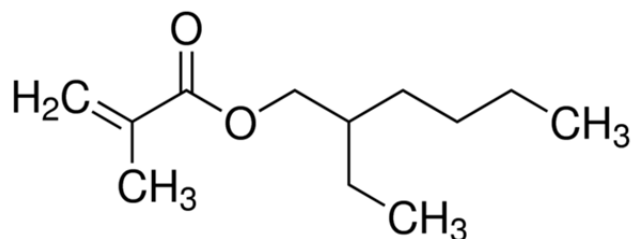
153. The drag reducers of Inaoka were prepared in a process in which a radical polymerization reaction is carried out in the presence of an oxygen absorbent, which prevents radicals from reacting with dissolved oxygen and being deactivated. Ex. 1007 at p. 3, lines 38-51. One of the stated benefits of this process is that because the radicals can be used solely for starting and maintaining the polymerization reaction, “it is possible to control the radical polymerization reaction with ease.” Ex. 1007 at p. 3, lines 49-51. The radical polymerization reaction is preferably an emulsion polymerization. Ex. 1007 at p. 4, lines 53-57. A person of ordinary skill in the art would have understood that a major benefit of using emulsion polymerization to prepare drag reducing polymers is that it provides for the production of high molecular weight polymers having a narrow distribution in individual chain molecular weights, also referred to as low dispersity. See for example Ex. 1013 at 351, 366; see also Ex. 1007 at p. 5, lines 2-13 (describing seeding, which is used to produce low dispersity).

154. Inaoka describes a number of preferred monomers that may be used for preparing the drag reducers, including (meth)acrylates represented by the formula  $\text{CH}_2 = \text{CR}_1\text{COOR}$  ( $\text{R}_1$  being H or  $\text{CH}_3$ , and R being a monovalent hydrocarbon group with a carbon number of 4-18, and more preferably 8-18). Ex. 1007 at p. 4, lines 25-27, 44-48. By selecting a compound in which R has a carbon number falling in the above range, “the compatibility of a fluid such as crude oil

and the high molecular weight polymer of the present invention is increased, and the effect as a drag reducer is improved.” Ex. 1007 at p. 4, lines 50-52.

155. Among the particularly preferred polymers is a 2-ethylhexyl methacrylate (2EHMA) homopolymer (in which R has a carbon number of 8). Ex. 1007 at p. 4, lines 48-50. Samples of the 2EHMA homopolymer are prepared in each of Examples 4, 5, 12, and 13. Ex. 1007 at pages 10-12. The 2EHMA homopolymer disclosed by Inaoka is the same polymer identified by the Lubrizol patents as Polymer A and which was shown by the Lubrizol patents to be an effective drag reducing agent in Albion Heavy Sour (AHS) crude oil and San Joaquin Valley Heavy (SJVH) crude oil. See Ex. 1001 at 13:12-36, 17:36-18:24; Ex. 1002 at 12:55-13:11, 17:1-56; Ex. 1003 at 12:52-13:10, 16:63-17:51; Ex. 1004 at 13:12-36, 17:31-18:18.

156. 2-ethylhexyl methacrylate – the monomeric constituent of the 2EHMA homopolymer – was known to contain oxygen heteroatoms. The chemical structure of the 2-ethylhexyl methacrylate monomer is shown below:



Accordingly, a person of ordinary skill in the art would have understood that a plurality of the repeat units in the 2EHMA homopolymer contain oxygen heteroatoms. Indeed, because 2EHMA is a homopolymer that is made up solely of 2-ethylhexyl methacrylate monomer segments, all of the repeat units of the 2EHMA homopolymer contain oxygen heteroatoms.

157. According to Inaoka, the high molecular weight polymers that are the subject of the patent application have a limiting viscosity value (represented by  $[\eta]$ ) not less than 5. Ex. 1007 at p. 7, lines 53-54. Inaoka describes the well-known relationship between limiting viscosity numbers and molecular weight known as the Kuhn-Mark-Houwink-Sakurada equation. Ex. 1007 at p. 7, lines 54-57. Although Inaoka explains that the molecular weight of a polymer cannot be determined in a straightforward manner from the limiting viscosity number, Inaoka teaches both (a) that a polymer with a limiting viscosity number of at least 5 is considered a high molecular weight polymer and (b) that such a polymer has a molecular weight of at least 5 million g/mol. Ex. 1007 at p. 7, line 57 to p. 8, line 4.

158. Inaoka also states that when adopting the polymer as a drag reducer, it is preferable that the polymer have a limiting viscosity value not less than 12. Ex. 1007 at p. 8, lines 5-12. Accordingly, a person of ordinary skill in the art would

have been led to prepare a 2EHMA polymer having a limiting viscosity value of not less than 12 for use as a drag reducer.

159. Using the information provided by Inaoka, a person of ordinary skill in the art would have been able to determine, at least within a reasonable range, the molecular weight of such a polymer. Specifically, such a person could determine, within a reasonable range, the molecular weight of such a polymer by using the Kuhn-Mark-Houwink-Sakurada equation provided by Inaoka, the disclosure that a  $[\eta]$  value of 5 is essentially equivalent to a molecular weight of 5 million, and the understanding that the constant “a” ranges between 0.5 and 0.8 for a good solvent, which would have been known by reference to standard polymer handbooks, for example, that provided in Ex. 1012. As shown in Attachment C, and using the information disclosed in Inaoka, I have calculated that a polymer of Inaoka having a limiting viscosity of 12 would have a molecular weight between about 14.9 million g/mol and 28.8 million g/mol.

160. Although Inaoka does not specify whether the disclosed molecular weight (of not less than 5 million g/mol) is a weight average molecular weight, a number average molecular weight, or a viscosity average molecular weight, a person of ordinary skill in the art would have understood that, in any case, a 2EHMA polymer having a limiting viscosity value of not less than 12 would have

a weight average molecular weight that is greater than the “at least  $1 \times 10^6$  g/mol” recited by certain claims of the Lubrizol patents.

161. For instance, if the molecular weight disclosed by Inaoka were a weight average molecular weight, then a polymer having a limiting viscosity value of not less than 12 would have a molecular weight of not less than 14.9 million g/mol, as calculated above, which is well above the “at least  $1 \times 10^6$  g/mol” recited by claims of the Lubrizol patents. Moreover, for a low dispersity polymer such as those generated by Inaoka using emulsion polymerization, a person of ordinary skill in the art would have understood that the weight average molecular weight of a 2EHMA sample would be not less than the viscosity average and number average molecular weights of the sample. See Ex. 1012 at 196-198. Therefore, if the molecular weight disclosed by Inaoka were either a viscosity average molecular weight or a number average molecular weight, then the minimum weight average molecular weight would be greater than 14.9 million g/mol.

162. A person of ordinary skill in the art also would have understood that a 2EHMA polymer having a limiting viscosity value of not less than 12 would contain “at least about 25,000 repeating units,” as recited by certain claims of the Lubrizol patents.

163. The number of repeat units in a homopolymer can easily be calculated by dividing the number average molecular weight of the homopolymer by the

molecular weight of a single repeat unit. A person of ordinary skill in the art would have been able to easily identify (either through conventional resources or by simple calculation) the molecular weight of the 2-ethylhexyl methacrylate repeat unit to be 198.30 g/mol.

164. A person of ordinary skill in the art would have understood that the number average molecular weight could be calculated by dividing the weight average molecular weight by the dispersity. For a low dispersity polymer such as that generated by Inaoka using emulsion polymerization, a person of ordinary skill in the art also would have understood that the number average molecular weight of a 2EHMA sample would be close to the weight average molecular weight of the sample. More particularly, a polymer such as that generated by Inaoka using emulsion polymerization would have been understood to have a dispersity between about 1.1 and 2.0. See Ex. 1013 at 365-366 (identifying the theoretical boundary of the dispersity as 2.0 for termination by coupling; note the theoretical boundary of the dispersity is 1.5 when termination occurs by disproportionation only). Therefore, based on a minimum weight average molecular weight of approximately 14.9 million g/mol, a person of ordinary skill in the art would have understood that the minimum number average molecular weight of such a 2EHMA polymer would be between about 7 million g/mol ( $14.9 \text{ million} / 2.0 \approx 7.4 \text{ million}$ ) and about 14 million g/mol ( $14.9 \text{ million} / 1.1 \approx 13.5 \text{ million}$ ).



165. By dividing the minimum number average molecular weight of the 2EHMA homopolymer by the molecular weight of a 2-ethylhexyl methacrylate repeat unit, a person of ordinary skill in the art would have easily calculated the minimum number of repeat units in the 2EHMA homopolymer to be between about 35,000 repeat units ( $7.0 \text{ million} / 198.30 \approx 35,300$  repeat units [dispersity of 2.0]) and about 68,000 repeat units ( $13.5 \text{ million} / 198.30 \approx 68,100$  repeat units [dispersity of 1.1]), which is well above the “at least about 25,000 repeating units” recited by certain claims of the Lubrizol patents.

166. Inaoka describes the polymers, including the preferred 2EHMA polymer, as being “suitably adopted in transporting of an organic liquid, such as crude oil through a conduit such as a pipeline.” Ex. 1007 at p. 17, lines 1-5.

167. While Inaoka does not describe the exact mechanism by which the drag reducing polymers function when added to crude oil flowing through a pipeline, a person of ordinary skill in the art would have understood that the drag reducing polymers of Inaoka achieve a reduction in frictional pressure losses associated with turbulent flow by suppressing the growth of turbulent eddies, that suppression being the mechanism by which drag reducing polymers were (and are) understood to function in turbulent flow by persons of ordinary skill in the art. See, for example, Ex. 1006 at 1:32-56, 6:67-7:15; see also Ex. 1001 at 1:28-37, Ex. 1002 at 1:33-42, Ex. 1003 at 1:33-42, Ex. 1004 at 1:33-42.

168. A person of ordinary skill in the art would have been led to add the 2EHMA drag reducing polymer to a liquid hydrocarbon at a concentration within the about 0.1 ppmw to about 500 ppmw range recited by certain claims of the Lubrizol patents. In particular, Inaoka specifically discloses that the amount of polymeric drag reducer added to a crude oil is preferably in a range of 0.1 ppm to 100 ppm, more preferably in a range of 0.5 ppm to 50 ppm. Ex. 1007 at p. 8, lines 18-20.

169. A person of ordinary skill in the art also would have understood that the addition of the 2EHMA drag reducing polymer to a liquid hydrocarbon would not cause a decrease in the viscosity of the liquid hydrocarbon.

170. As a general matter, a person of ordinary skill in the art would have understood that the addition of polymers having the high molecular weights disclosed by Inaoka would not cause a reduction in the viscosity of a liquid hydrocarbon. As shown by the Holtmyer Patent, for example, adding increasing amounts of a high molecular weight polymeric drag reducing agent to a kerosene liquid hydrocarbon actually results in an *increase* in the viscosity of the treated liquid hydrocarbon. Ex. 1006 at Table VI. Accordingly, a person of ordinary skill in the art would have understood that addition of the high molecular weight drag reducing polymers of Inaoka to a liquid hydrocarbon such as that defined by the claims of the Lubrizol patents would produce a treated liquid hydrocarbon in

which the viscosity of the treated liquid hydrocarbon is not less than the viscosity of the liquid hydrocarbon prior to treatment with the drag reducing polymer.

171. Further, the high molecular weight polymers of Inaoka are described as being “directly used as a drag reducer.” Ex. 1007 at p. 8, lines 15-16. According to Inaoka, the drag reducer containing the high molecular weight polymer may *optionally* contain a known adduct, such as an organic solvent. Ex. 1007 at p. 8, lines 16-18. On the other hand, for example, the high molecular weight polymer may be used by extracting the polymer, *i.e.* by removing water. Ex. 1007 at p. 8, lines 26-29. It would have been obvious to a person of ordinary skill in the art to have introduced the drag reducing 2EHMA polymer into a crude oil either alone or with a relatively small amount of solvent, such that addition of the drag reducer would not lower the viscosity of the crude oil.

## **XI. The Combination of Inaoka and Carnahan**

172. Inaoka does not explicitly disclose the introduction of the 2EHMA drag reducing polymer into a pipeline containing a flow of a crude oil having an asphaltene content of at least 3 weight percent and an API gravity of less than 26°, as recited by the claims of the Lubrizol patents. However, it would have been obvious to a person of ordinary skill in the art to have introduced the 2EHMA drag reducing polymer disclosed by Inaoka into a pipeline containing a flow of crude oil having this combination of properties.

173. Crude oils containing an asphaltene content of at least 3 weight percent and an API gravity of less than 26° were well-known in 2005. See, for example, Ex. 1017 at 12, Table I (identifying Alaska-93, Alaska-95, Schuricht, and Heavy California crude oils); Ex. 1018 at 557-558 (identifying Prudhoe Bay Alaskan crude oil); see also the Lubrizol patents, Exs. 1001-1004, at Table 1 (identifying a number of crude oils).

174. It also was well understood that there was (and is) a significant economic advantage to reducing the frictional pressure losses that occur during the transport of crude oils through pipelines. For instance, the Holtmyer Patent cited by Inaoka as conveying the state of the art explained that “[i]n order to compensate for the frictional pressure loss encountered in the turbulent flow of such hydrocarbon liquids, considerable energy generally in the form of pumping horsepower must be expended. Thus, reduction of the frictional pressure loss in the flow of such hydrocarbon liquids brings about an advantageous reduction in horsepower requirements, or alternatively, an increased flow rate of the hydrocarbon liquids under the same pumping conditions.” Ex. 1006 at 1:56-65; see also Ex. 1001 at 1:15-30, Ex. 1002 at 1:20-35, Ex. 1003 at 1:20-35, Ex. 1004 at 1:20-35 (describing problems associated with frictional pressure drop and stating that drag reducing additives are commonly used in the industry to alleviate the problems).

175. A person of ordinary skill in the art would have recognized that these advantages would be realized in the transport of crude oil in general, which would include the “heavy” crude oil recited in the claims of the Lubrizol patents.

176. Accordingly, a person of ordinary skill in the art would have been motivated to introduce the 2EHMA drag reducing polymer disclosed by Inaoka into a pipeline containing a flow of crude oil having the claimed asphaltene content and API gravity properties to obtain the well-known economic benefits associated with drag reduction.

177. Moreover, a person of ordinary skill in the art would have had a reasonable expectation that the 2EHMA drag reducing polymer of Inaoka would be effective at reducing the frictional pressure losses associated with turbulent flow when introduced into a crude oil having an asphaltene content of at least 3 weight percent and an API gravity of less than 26°, as recited by the claims of the Lubrizol patents.

178. Inaoka states that the 2EHMA polymer has each of the properties that were generally understood as being required for effective drag reduction; namely, a high molecular weight, a straight-chain structure with minimal branching, and solubility. See Ex. 1007 at Abstract. Specifically, the testing performed by Inaoka demonstrates that the 2EHMA polymer is soluble in an organic solvent (specifically tetrahydrofuran). Ex. 1007 at p. 9, lines 10-14; p. 13 (Table 1).

However, Inaoka does not provide any test results demonstrating the solubility (or effectiveness) of the 2EHMA polymer in a crude oil, such as a crude oil having an asphaltene content of at least 3 weight percent and an API gravity of less than 26°.

179. Because the effectiveness of a drag reducing agent in a liquid hydrocarbon was known to be heavily dependent on the solubility of the drag reducing agent in that liquid hydrocarbon (see, e.g., ¶¶29, 34), a person of ordinary skill in the art would have understood that the effectiveness of the 2EHMA drag reducing polymer in a target crude oil, such as a crude oil having an asphaltene content of at least 3 weight percent and an API gravity of less than 26°, would depend on the solubility of the 2EHMA drag reducing polymer in that crude oil. Therefore, a person of ordinary skill in the art contemplating the use of the 2EHMA drag reducing polymer of Inaoka in a target crude oil having an asphaltene content of at least 3 weight percent and an API gravity of less than 26° would have sought to evaluate the solubility of the 2EHMA drag reducing polymer in such a crude oil.

180. To evaluate the solubility of a drag reducing polymer in a particular liquid hydrocarbon such as a crude oil, a person of ordinary skill in the art would have understood that the solubility could either be tested by experimental methods or be predicted by comparing the solubility parameters of the two materials. Solubility parameters were (and are) commonly consulted in order to predict the

solubility of one material in another without the need for time-consuming, and sometimes expensive, experimentation. Therefore, to easily evaluate the solubility of the 2EHMA drag reducing polymer in a crude oil having at least 3 weight percent asphaltene and an API gravity of less than about 26°, a person of ordinary skill in the art would have sought to compare the solubility parameters of the drag reducing polymer and the crude oil.

181. A person of ordinary skill in the art would have easily been able to identify the solubility parameter of the 2EHMA polymer of Inaoka. First, the solubility parameters of many common polymers were published in well-known handbooks, such as the *Polymer Handbook* referenced by the Lubrizol patents (i.e., Brandrup). Ex. 1011 at 702-711; Table 10. Additionally, it was well understood in 2005 that one could easily calculate the solubility parameter of a polymer using the group contribution method detailed in Brandrup. Ex. 1011 at 686-687; Table 4, 5.

182. I have calculated a value for the solubility parameter of the 2EHMA polymer of Inaoka using the group contribution method presented in Brandrup at B11 and C30 (Ex. 1011 at 677, 683), i.e., the equations recited in the Lubrizol patents, as shown in Attachment D, and arrived at a solubility parameter value of about 18.04 MPa<sup>1/2</sup> (which corresponds with the value provided by the Lubrizol

patents for the 2EHMA polymer, see Ex. 1001 at 13:12-36; Ex. 1002 at 12:55-13:11; Ex. 1003 at 12:52-13:10; Ex. 1004 at 13:12-36).

183. One of the variables needed for calculating the solubility parameter of a polymer using the group contribution method of Brandrup is the molar volume of the polymer in question. Because neither the experimental molar volume nor the density (from which the molar volume could be calculated with knowledge of the compound's molecular weight) of the 2EHMA polymer is readily available from reliable sources, I calculated the molar volume of the 2EHMA polymer using the group contribution method described in Brandrup. Ex. 1011 at 682-683; Table 3. This calculation also is shown in Attachment D.

184. A person of ordinary skill in the art also would have been able to generally identify the solubility parameter of a crude oil having an asphaltene content of at least 3 weight percent and an API gravity of less than about 26°. A person of ordinary skill in the art would have recognized that, due to the complexity of crude oil, it would be very difficult to calculate the solubility parameter of such a crude oil using the group contribution methods of Brandrup. Therefore, a person of ordinary skill in the art would have understood that the solubility parameter of such a crude oil must be determined using other methods.

185. Because crude oil is a well-studied fluid, a person of ordinary skill in the art would have sought to locate the solubility parameters of crude oils having



asphaltene contents of at least 3 weight percent and API gravities of less than about 26° within the published literature on the subject. A person of ordinary skill in the art would have expected that studies on the solubility (and solubility parameters) of crude oil (and crude oil components) had been performed because of the importance of maintaining the heavier components of crude oil, such as asphaltenes, dispersed/solubilized within the crude oil during transport (precipitation of those components within a conduit can severely restrict the flow of crude oil, which is highly undesirable).

186. The book titled *Asphaltenes and Asphalts*, 2, published in 2000 as part of an Elsevier Science B.V. series related to Developments in Petroleum Science, contains a chapter titled “Precipitation of Asphaltenes in Heavy Oil and Tar Sands” written by Norman F. Carnahan (Ex. 1008; “Carnahan”). Carnahan generally describes the study of asphaltene solubility in crude oils, including through solubility parameter models. Because of its subject matter and its publication prior to 2005, a person of ordinary skill in the art would have looked to Carnahan as a potential source of solubility parameters for heavy, asphaltenic crude oils.

187. Indeed, having disclosed the solubility parameter of asphaltenes as being about 20 MPa<sup>1/2</sup> (Ex. 1008 at 324), Carnahan continues with a section titled “Solubility parameters of petroleum fluids.” Ex. 1008 at 325. Under this heading Carnahan states:

“Solubility parameter values for light crude components depend on composition, and are often in the range of about 6-8 hildebrands. . . . Solubility parameter values for medium and heavy oils are greater than those for light oils. Only a limited amount of reliable experimental data has been reported, although none for asphaltene phase behavior in heavy oils and tar sands. The expected value of the solubility parameter of such heavy petroleum fluids is about 8-10 hildebrands.”

Ex. 1008 at 325. Hildebrand units ( $[\text{cal}/\text{cm}^3]^{1/2}$ ) are readily converted to  $\text{MPa}^{1/2}$  units by multiplying by 2.046 (i.e.  $1 [\text{cal}/\text{cm}^3]^{1/2} = 2.046 \text{ MPa}^{1/2}$ ). Accordingly, Carnahan teaches a person of ordinary skill in the art that the solubility parameters of heavy, asphaltenic crude oils are expected to fall within the range of about 16.4  $\text{MPa}^{1/2}$  to 20.5  $\text{MPa}^{1/2}$ .

188. Carnahan’s teaching is supported by the article titled *Asphaltene Precipitation And Solvent Properties of Crude Oils*, written by J.S. Buckley et al. and published in *Petroleum Science and Technology*, vol. 16, no. 3-4, in 1998 (Ex. 1017; “Buckley”). Buckley teaches that the solubility parameter of a crude oil can be estimated by using the refractive index of that crude oil. Because of its subject matter and its publication prior to 2005, a person of ordinary skill in the art also would have looked to Buckley as a potential source of solubility parameters for asphaltenic crude oils.

189. Specifically, Buckley suggests that the solubility parameters of hydrocarbons have a linear relationship with a function of the refractive indexes of the hydrocarbons:  $F_{RI} = (n^2 - 1)/(n^2 + 2)$ . [Note that “n” is the standard abbreviation for refractive index; see also Ex. 1017 at 28 (identifying the symbol “n” as refractive index).] This relationship is shown graphically in Figure 1 of Buckley. Ex. 1017 at 4. Buckley also experimentally measured the refractive indexes (RI) of a number of heavy, asphaltenic crude oils. Ex. 1017 at 12 (Table I). By substituting the RI values for those oils into the above equation and plotting the results in Figure 1, I was able to estimate the solubility parameters of the following crude oils using the teachings of Buckley:

- A-93 crude oil (API gravity 25.5°; n-C<sub>5</sub> asphaltenes 10.9%):

$$RI = 1.522; F_{RI} \approx 0.305; \text{solubility parameter} \approx 18 \text{ MPa}^{1/2}$$

- A-95 crude oil (API gravity 25.2°; n-C<sub>5</sub> asphaltenes 5.0%):

$$RI = 1.518; F_{RI} \approx 0.303; \text{solubility parameter} \approx 18 \text{ MPa}^{1/2}$$

- Schuricht crude oil (API gravity 24.6°; n-C<sub>5</sub> asphaltenes 9.9%):

$$RI = 1.514; F_{RI} \approx 0.301; \text{solubility parameter} \approx 18 \text{ MPa}^{1/2}$$

- Heavy CA crude oil (API gravity 19.0°; n-C<sub>5</sub> asphaltenes 6.8%):

$$RI = 1.565; F_{RI} \approx 0.326; \text{solubility parameter} \approx 20 \text{ MPa}^{1/2}$$

190. Accordingly, a person of ordinary skill in the art would have understood from Buckley that the solubility parameters of crude oils having an

asphaltene content of at least 3 weight percent and an API gravity less than 26° were expected to fall squarely within the range taught by Carnahan.

191. Because the solubility parameter of the 2EHMA polymer of Inaoka (18.04 MPa<sup>1/2</sup>) falls squarely within the range taught by Carnahan (about 16.4 MPa<sup>1/2</sup> to 20.5 MPa<sup>1/2</sup>), a person of ordinary skill in the art would have had a reasonable expectation that the 2EHMA polymer would be soluble in a heavy crude oil, such as one having an asphaltene content of at least 3 weight percent and an API gravity less than about 26°.

192. Because 2EHMA (a) is disclosed by Inaoka as having a straight-chain structure with minimal branching and a sufficiently high molecular weight to be suitable as a drag reducer and (b) would have been expected by a person of ordinary skill in the art to be soluble in heavy, asphaltenic crude oil, a person of ordinary skill in the art would have had a reasonable expectation that the 2EHMA drag reducing polymer would be effective at achieving drag reduction in a crude oil having an asphaltene content of at least 3 weight percent and an API gravity of less than about 26° undergoing turbulent flow through a pipeline.

## **XII. The Combination of Inaoka and Strausz**

193. Inaoka does not explicitly disclose the introduction of the 2EHMA drag reducing polymer into a pipeline containing a flow of a crude oil having an asphaltene content of at least 3 weight percent and an API gravity of less than 26°,

as recited by the claims of the Lubrizol patents. However, it would have been obvious to a person of ordinary skill in the art to have introduced the 2EHMA drag reducing polymer disclosed by Inaoka into a pipeline containing a flow of crude oil having the recited combination of properties.

194. Crude oils containing an asphaltene content of at least 3 weight percent and an API gravity of less than 26° were well-known in 2005. See, for example, Ex. 1017 at 12, Table I (identifying Alaska-93, Alaska-95, Schuricht, and Heavy California crude oils); Ex. 1018 at 557-558 (identifying Prudhoe Bay Alaskan crude oil); see also the Lubrizol patents, Exs. 1001-1004, at Table 1 (identifying a number of crude oils).

195. It also was well understood that there was (and is) a significant economic advantage to reducing the frictional pressure losses that occur during the transport of crude oils through pipelines. For instance, the Holtmyer Patent cited by Inaoka as conveying the state of the art explained that “[i]n order to compensate for the frictional pressure loss encountered in the turbulent flow of such hydrocarbon liquids, considerable energy generally in the form of pumping horsepower must be expended. Thus, reduction of the frictional pressure loss in the flow of such hydrocarbon liquids brings about an advantageous reduction in horsepower requirements, or alternatively, an increased flow rate of the hydrocarbon liquids under the same pumping conditions.” Ex. 1006 at 1:56-65;

see also Ex. 1001 at 1:15-30, Ex. 1002 at 1:20-35, Ex. 1003 at 1:20-35, Ex. 1004 at 1:20-35 (describing problems associated with frictional pressure drop and stating that drag reducing additives are commonly used in the industry to alleviate the problems).

196. A person of ordinary skill in the art would have recognized that these advantages would be realized in the transport of crude oil in general, which would include the “heavy” crude oil recited in the claims of the Lubrizol patents.

197. Accordingly, a person of ordinary skill in the art would have been motivated to introduce the 2EHMA drag reducing polymer disclosed by Inaoka into a pipeline containing a flow of crude oil having the recited asphaltene content and API gravity properties to obtain the well-known economic benefits associated with drag reduction.

198. Moreover, a person of ordinary skill in the art would have had a reasonable expectation that the 2EHMA drag reducing polymer of Inaoka would be effective at reducing the frictional pressure losses associated with turbulent flow when introduced into a crude oil having an asphaltene content of at least 3 weight percent and an API gravity of less than 26°, as recited by the claims of the Lubrizol patents.

199. Inaoka states that the 2EHMA polymer has each of the properties that were generally understood as being required for effective drag reduction; namely, a

high molecular weight, a straight-chain structure with minimal branching, and solubility. See Ex. 1007 at Abstract. Specifically, the testing performed by Inaoka demonstrates that the 2EHMA polymer is soluble in an organic solvent (specifically tetrahydrofuran). Ex. 1007 at p. 9, lines 10-14; p. 13 (Table 1). However, Inaoka does not provide any test results demonstrating the solubility (or effectiveness) of the 2EHMA polymer in a crude oil, such as a crude oil having an asphaltene content of at least 3 weight percent and an API gravity of less than 26°.

200. Because the effectiveness of a drag reducing agent in a liquid hydrocarbon was known to be heavily dependent on the solubility of the drag reducing agent in that liquid hydrocarbon (see, e.g., ¶¶29, 34), a person of ordinary skill in the art would have understood that the effectiveness of the 2EHMA drag reducing polymer in a target crude oil, such as a crude oil having an asphaltene content of at least 3 weight percent and an API gravity of less than 26°, would depend on the solubility of the 2EHMA drag reducing polymer in that crude oil. Therefore, a person of ordinary skill in the art contemplating the use of the 2EHMA drag reducing polymer of Inaoka in a crude oil having an asphaltene content of at least 3 weight percent and an API gravity of less than 26° would have sought to evaluate the solubility of the 2EHMA drag reducing polymer in such a crude oil.

201. To evaluate the solubility of a drag reducing polymer in a particular liquid hydrocarbon, such as a crude oil, a person of ordinary skill in the art would have understood that the solubility could either be tested by experimental methods or be predicted by comparing the solubility parameters of the two materials. Solubility parameters were (and are) commonly consulted in order to predict the solubility of one material in another without the need for time-consuming, and sometimes expensive, experimentation. Therefore, to easily evaluate the solubility of the 2EHMA drag reducing polymer in a crude oil having at least 3 weight percent asphaltene and an API gravity of less than about 26°, a person of ordinary skill in the art would have sought to compare the solubility parameters of the drag reducing polymer and the crude oil.

202. A person of ordinary skill in the art would have easily been able to identify the solubility parameter of the 2EHMA polymers of Inaoka. First, the solubility parameters of many common polymers were published in well-known handbooks, such as the *Polymer Handbook* referenced by the Lubrizol patents (i.e., Brandrup). Ex. 1011 at 702-711; Table 10. Additionally, it was well understood in 2005 that one could easily calculate the solubility parameter of a polymer using the group contribution method presented in Brandrup. Ex. 1011 at 686-687; Table 4, 5.



203. I have calculated a value for the solubility parameter of the 2EHMA polymer of Inaoka using the group contribution method presented in Brandrup at B11 and C30 (Ex. 1011 at 677, 683), i.e., the equations recited in the Lubrizol patents, as shown in Attachment D, and arrived at a solubility parameter value of about  $18.04 \text{ MPa}^{1/2}$  (which corresponds with the value provided by the Lubrizol patents for the 2EHMA polymer, see Ex. 1001 at 13:12-36; Ex. 1002 at 12:55-13:11; Ex. 1003 at 12:52-13:10; Ex. 1004 at 13:12-36).

204. One of the variables needed for calculating the solubility parameter of a polymer using the group contribution method of Brandrup is the molar volume of the polymer in question. Because neither the experimental molar volume nor the density (from which the molar volume could be calculated with knowledge of the compound's molecular weight) of the 2EHMA polymer is readily available from reliable sources, I calculated the molar volume of the 2EHMA polymer using the group contribution method described in Brandrup. Ex. 1011 at 682-683; Table 3. This calculation also is shown in Attachment D.

205. A person of ordinary skill in the art also would have been able to generally determine a range of solubility parameters for a crude oil having an asphaltene content of at least 3 weight percent and an API gravity of less than about  $26^\circ$ . A person of ordinary skill in the art would have recognized that, due to the complexity of crude oil, it would be very difficult to calculate the solubility

parameter of such a crude oil using the group contribution methods of Brandrup.

Therefore, a person of ordinary skill in the art would have understood that the solubility parameter of such a crude oil must be determined using other methods.

206. Crude oil is a complex fluid containing a number of different components. When faced with determining the solubility parameter of such a fluid, a person of ordinary skill in the art would have looked to solubility characteristics of its individual components to determine a range within which the solubility parameter of the crude oil falls.

207. In particular, a person of ordinary skill in the art would start by looking at the solubilized and dispersed components of crude oil. The solubilized and dispersed constituents of crude oil include resins, asphaltenes, and cokes. By determining the range of fluids (that have known or easily calculated solubility parameters) that are capable of solubilizing or dispersing these crude oil components, a person of ordinary skill in the art would be able to obtain a range of solubility parameters within which a heavy crude oil—in which these constituents are solubilized or dispersed in relatively high amounts compared to a light crude oil—would fall.

208. To obtain a (relatively) narrow range within which a solubility parameter of a heavy crude oil falls, a person of ordinary skill in the art would have looked toward the solubility characteristics of the higher molecular weight

components of crude oil, such as asphaltene. The rationale is as follows: as the molecular weight of the components increases, the range of solubility parameters for fluids capable of solubilizing (dispersing) these higher molecular weight components narrows. Therefore, looking to the solubility characteristics of the higher molecular weight components, such as asphaltene, would allow a person of ordinary skill in the art to refine significantly the range for the solubility parameter of a heavy crude oil. Note that one would have been deterred from performing this analysis on the highest molecular weight constituents of crude oil, i.e. cokes, because they would be very difficult to solubilize (disperse) in easily studied fluids.

209. A person of ordinary skill in the art also would have been familiar with this type of experimentation. This type of experimentation is (was) often referred to as solvent fractionation. Among other things, it was used to refine the solubility parameter value of a complex or unknown material.

210. The book titled *The Chemistry of Alberta Oil Sands, Bitumens and Heavy Oils*, was written by Otto P. Strausz and Elizabeth M. Lown, and published by Alberta Energy Research Institute in 2003 (Ex. 1009; “Strausz”). Strausz addresses a number of topics related to the composition and properties of crude oils. In Chapter 14, Strausz describes the properties of asphaltene, including its solubility characteristics. Ex. 1009 at 464. Because of its subject matter and its

publication prior to 2005, a person of ordinary skill in the art looking to determine the solubility parameter of a heavy, asphaltenic crude oil would have been led to Strausz.

211. Strausz generally describes the relationship between the molecular weight of the components of crude oil and the range of fluids that are capable of solubilizing each component. Specifically, Strausz states “solvents for coke represent a subset of solvents for asphaltene, asphaltene for resins, resins for aromatics, meaning that solvents for asphaltene are solvents for resins, etc....” Ex. 1009 at 473. This relationship is also shown graphically by Strausz. See Ex. 1009 at 474 (note the narrowing range of fluids in which the component is soluble as one moves from the lower molecular weight aromatics to the intermediate molecular weight resins and on to the higher molecular weight asphaltenes).

212. Although Strausz states that asphaltene forms a colloidal system in crude oil, and would therefore technically be “dispersed” rather than “soluble” in crude oil, Strausz recognizes that the solubility parameter model is nevertheless applicable in such a system. Accordingly, Strausz generally describes the ability of a liquid, such as crude oil, to disperse asphaltene in terms of “solubility”. See Ex. 1009 at 464 (“The solubility of asphaltene in the maltene of crude oil and the stability of the asphaltene solution with respect to precipitation are of considerable

interest to all phases of the petroleum industry....”). For consistency, I have used the same terminology as used by Strausz.

213. Strausz describes the solubility characteristics of the high molecular weight asphaltenes in terms of solubility parameter. In particular, Strausz describes a study of the correlation between the solubility of asphaltene in a variety of test solvents and the solubility parameter of each of those test solvents. Ex. 1009 at 467-468. The study uses the equation  $\delta_2 = [(\Delta H_v - RT)/V]^{1/2}$  for calculating the solubility parameters of the test solvents. Ex. 1009 at 468, Table 14.1. That study demonstrated that “the solvation energy of the solvents with  $\delta_2$  in the 17.1-22.1 MPa<sup>1/2</sup> range is sufficiently large to overcome the cohesion energy of asphaltene and cause solubilization.” Ex. 1009 at 467. Therefore, Strausz teaches that to solubilize the typical asphaltene contents of heavy crude oils, a hydrocarbon fluid should have a solubility parameter between about 17.1 MPa<sup>1/2</sup> and about 22.1 MPa<sup>1/2</sup> (as determined using the above equation).

214. A person of ordinary skill in the art would have understood from Strausz that, generally, in order for a heavy crude oil flowing through a pipeline to have had an asphaltene content of at least 3 weight percent, that crude oil would have a solubility parameter within the disclosed range of about 17.1 MPa<sup>1/2</sup> to about 22.1 MPa<sup>1/2</sup>.

215. Because the solubility parameter of the 2EHMA polymer of Inaoka ( $18.04 \text{ MPa}^{1/2}$ ) falls squarely within the range taught by Strausz ( $17.1 \text{ MPa}^{1/2}$  and  $22.1 \text{ MPa}^{1/2}$ ), a person of ordinary skill in the art would have had a reasonable expectation that the 2EHMA polymer would be soluble in a crude oil having an asphaltene content of at least 3 weight percent and an API gravity less than about  $26^\circ$ .

216. Because 2EHMA (a) is disclosed by Inaoka as having a straight-chain structure with minimal branching and a sufficiently high molecular weight to be suitable as a drag reducer and (b) would have been expected by a person of ordinary skill in the art to be soluble in a heavy, asphaltenic crude oil, a person of ordinary skill in the art would have had a reasonable expectation that the 2EHMA drag reducing polymer would be effective at achieving drag reduction in a crude oil having an asphaltene content of at least 3 weight percent and an API gravity of less than about  $26^\circ$  undergoing turbulent flow through a pipeline.

217. Further, based on the teaching of Strausz, including the table providing solubility parameters for many of the components of crude oil (Ex. 1009 at 468, Table 14.1), a person of ordinary skill in the art would have also understood that at least some, and likely a fairly large proportion, of the crude oils having an API gravity of less than about  $26^\circ$  and an asphaltene content of at least 3 weight percent would have solubility parameters between about  $17.1 \text{ MPa}^{1/2}$  (the lower

bound of the range disclosed by Strausz, which is within  $1 \text{ MPa}^{1/2}$  of the solubility parameter of the 2EHMA polymer) and  $22.04 \text{ MPa}^{1/2}$  (which is within  $4 \text{ MPa}^{1/2}$  of the solubility parameter of the 2EHMA polymer). As such, a person of ordinary skill in the art would have understood that at least some, and likely a fairly large proportion, of these crude oils would have solubility parameters within  $4 \text{ MPa}^{1/2}$  of the solubility parameter of the 2EHMA polymer.

218. Similarly, a person of ordinary skill in the art would have understood that at least some, and likely a fairly large proportion, of the crude oils having an API gravity of less than about  $26^\circ$  and an asphaltene content of at least 3 weight percent would have solubility parameters between  $17.1 \text{ MPa}^{1/2}$  (the lower bound of the range disclosed by Strausz, which is within  $1 \text{ MPa}^{1/2}$  of the solubility parameter of the 2EHMA polymer) and  $20.54 \text{ MPa}^{1/2}$  (which is within  $2.5 \text{ MPa}^{1/2}$  of the solubility parameter of the 2EHMA polymer). A person of ordinary skill in the art would therefore have understood that at least some, and likely a fairly large proportion, of these crude oils would have solubility parameters within  $2.5 \text{ MPa}^{1/2}$  of the solubility parameter of the 2EHMA polymer.

219. In general, it would have been obvious in 2005 for a person of ordinary skill in the art to select a drag reducing polymer for a crude oil in which the drag reducing polymer was expected to be most effective, i.e., to provide the highest degree of drag reduction. It was well known in the art that the closer the

solubility parameter of a drag reducing polymer is to that of a liquid hydrocarbon, the more soluble one would have expected the polymer to be in that hydrocarbon, and thus the higher the degree of drag reduction that would have been expected. To achieve the greatest drag reduction, therefore, it would have been obvious for one of ordinary skill in the art to have selected a drag reducing polymer for use in a crude oil in which it was expected to be highly soluble, i.e., a crude oil having a close solubility parameter. Indeed, selecting a drag reducing polymer that would produce the greatest drag reducing effect would have been a matter of routine optimization.

220. Accordingly, it would have been obvious for a person of ordinary skill in the art to have utilized the 2EHMA polymer of Inaoka in a crude oil having a solubility parameter within  $4 \text{ MPa}^{1/2}$ , and more preferably within  $2.5 \text{ MPa}^{1/2}$ , of the solubility parameter of the 2EHMA drag reducing polymer.

221. By reciting that the solubility parameters of the drag reducing agent and the liquid hydrocarbon are close to one another, the Lubrizol patents do no more than use solubility parameters (in a conventional manner) to describe the already well-known relationship between the solubility of a drag reducing polymer (such as those discussed herein) in a fluid and the expected effectiveness of the drag reducing polymer in that fluid. In fact, Lubrizol's selection of  $4 \text{ MPa}^{1/2}$  as the claimed range corresponds with Strausz's description that, typically, materials



whose solubility parameters differ by greater than  $4 \text{ MPa}^{1/2}$  are largely immiscible or insoluble. Ex. 1009 at 466 (“liquids differing by two hildebrands [i.e., about  $4 \text{ MPa}^{1/2}$ ] in their [solubility parameter] values are generally incompletely miscible . . .”).

### **XIII. Conclusion**

222. I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code.



Date: 10/05/2016

Thomas H. Epps, III, Ph.D.

# **ATTACHMENT A**

# iDma-Holtmyer Molecular Weight Calculation

Table 9  $\rightarrow$  intrinsic viscosity  $\rightarrow 5.5 \text{ dl/g} = \langle \eta \rangle$

equation on page 474:  $\langle \eta \rangle = 8.64 \times 10^{-5} M_v^{0.64}$   
for methacrylates in butyl acetate at  $23^\circ\text{C}$

$$5.5 = 8.64 \times 10^{-5} M_v^{0.64}$$

$$6.36 \times 10^4 = M_v^{0.64}$$

$$\underline{32.1 \times 10^6 \text{ g/mol} = M_v}$$

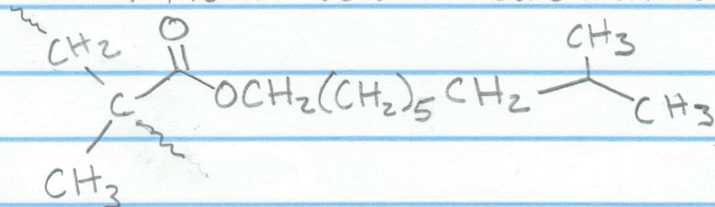
or

$$M_v \approx 32 \text{ million g/mol}$$

# **ATTACHMENT B**



iDMA molar volume calculation - group contribution



Using group contribution molar volume obtained from  
Brandrup VII/685, Table 3

$$\text{CH}_3 \text{ at } 33.5 \text{ cm}^3/\text{mol} \rightarrow 3 \times 33.5 = 100.5 \text{ cm}^3/\text{mol}$$

$$\text{CH}_2 \text{ at } 16.1 \text{ cm}^3/\text{mol} \rightarrow 8 \times 16.1 = 128.8 \text{ cm}^3/\text{mol}$$

$$\text{CH} \text{ at } -1.0 \text{ cm}^3/\text{mol} \rightarrow 1 \times -1.0 = -1.0 \text{ cm}^3/\text{mol}$$

$$\text{C} \text{ at } -19.2 \text{ cm}^3/\text{mol} \rightarrow 1 \times -19.2 = -19.2 \text{ cm}^3/\text{mol}$$

$$-\text{COO}- \text{ at } 18.0 \text{ cm}^3/\text{mol} \rightarrow 1 \times 18.0 = 18.0 \text{ cm}^3/\text{mol}$$

$$\boxed{227.1 \text{ cm}^3/\text{mol}}$$



iDMA solubility parameter - group contribution

$$\bar{V}(\text{molar volume}) = 227.1 \text{ cm}^3/\text{mol}$$

$$\text{units} \rightarrow 3 \times \text{CH}_3 + 8 \times \text{CH}_2 + 1 \times \text{CH} + 1 \times \text{C} + 1 \times \text{COO}$$

$$F_{di} = 3 \times 420 + 8 \times 270 + 1 \times 80 + 1 \times -70 + 1 \times 390$$

$$F_{di} = 3820 \text{ J}^{1/2} \text{ cm}^{3/2} \text{ mol}^{-1}$$

$$F_{pi} = 1 \times 490 = 490 \text{ J}^{1/2} \text{ cm}^{3/2} \text{ mol}^{-1} \text{ from } -\text{COO}-$$

$$E_{hi} = 1 \times 7000 = 7000 \text{ J/mol from } -\text{COO}-$$

$$\sigma = \left( \sigma_d^2 + \sigma_p^2 + \sigma_h^2 \right)^{1/2} = \left[ (16.82)^2 + (2.16)^2 + (5.55)^2 \right]^{1/2}$$

$$\sigma = 17.84 \text{ MPa}^{1/2}$$

# **ATTACHMENT C**



(1 of 2)

2EHMA - Inaoka Molecular Weight Calculation  
limiting viscosity number  $\rightarrow [\eta]$

Kuhn-Mark-Houwink-Sakurada equation  $\rightarrow [\eta] = K_m M^a$

note: this should be Houwink, but following  
typo in patent on page 7

$K_m \rightarrow \text{constant}$  ;  $a \rightarrow \text{constant}$

for polymer in Inaoka (page 8):

$$5 = K_m (5 \times 10^6)^a \quad \text{as a lower bound}$$

for drag reduction (page 8)

$$[\eta] \geq 12 \quad \text{or} \quad 12 = K_m M^a \quad \text{as lower bound}$$

$0.5 < a < 0.8$  for polymer dissolved in theta  
solvent (0.5) to good solvent (0.8) for flexible  
coil chains (see Young & Lovell; page 197)

$$[\eta] = 5 = K_m (5 \times 10^6)^a \quad \text{and} \quad [\eta] = 12 = K_m M^a$$

$$\frac{12}{5} = \frac{K_m M^a}{K_m (5 \times 10^6)^a} \Rightarrow 2.4 = \frac{M^a}{(5 \times 10^6)^a}$$



(2 of 2)

for  $a = 0.8$

$$2.4 = \frac{M^{0.8}}{(5 \times 10^6)^{0.8}} \Rightarrow M^{0.8} = 5.49 \times 10^5$$

$\Downarrow$

$$M = 14.9 \times 10^6 \text{ molecular weight}$$

for  $a = 0.5$

$$2.4 = \frac{M^{0.5}}{(5 \times 10^6)^{0.5}} \Rightarrow M^{0.5} = 5.37 \times 10^3$$

$\Downarrow$

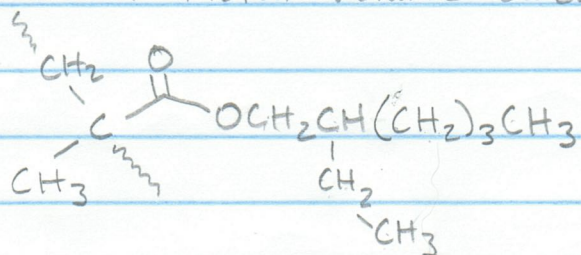
$$M = 28.8 \times 10^6 \text{ molecular weight}$$

$$\text{for } [\eta] = 12 \Rightarrow 14.9 \times 10^6 \leq M \leq 28.8 \times 10^6$$

assuming units of Daltons or g/mol

# **ATTACHMENT D**

2EHMA molar volume calculation - group contribution



Using group contribution molar volume obtained from  
Brandrup VII/685, Table 3

$$\text{CH}_3 \text{ at } 33.5 \text{ cm}^3/\text{mol} \rightarrow 3 \times 33.5 = 100.5 \text{ cm}^3/\text{mol}$$

$$\text{CH}_2 \text{ at } 16.1 \text{ cm}^3/\text{mol} \rightarrow 6 \times 16.1 = 96.6 \text{ cm}^3/\text{mol}$$

$$\text{CH} \text{ at } -1.0 \text{ cm}^3/\text{mol} \rightarrow 1 \times -1.0 = -1.0 \text{ cm}^3/\text{mol}$$

$$\text{C} \text{ at } -19.2 \text{ cm}^3/\text{mol} \rightarrow 1 \times -19.2 = -19.2 \text{ cm}^3/\text{mol}$$

$$-\text{COO}- \text{ at } 18.0 \text{ cm}^3/\text{mol} \rightarrow 1 \times 18.0 = 18.0 \text{ cm}^3/\text{mol}$$

$$\boxed{194.9 \text{ cm}^3/\text{mol}}$$



2EHMA solubility parameter calculation - group contribution

$$\bar{V}(\text{molar volume}) = 194.9 \text{ cm}^3/\text{mol}$$

$$\text{units} \rightarrow 3 \times \text{CH}_3 + 6 \times \text{CH}_2 + 1 \times \text{CH} + 1 \times \text{C} + 1 \times \text{COO}$$

$$F_{di} = 3 \times 420 + 6 \times 270 + 1 \times 80 + 1 \times -70 + 1 \times 390$$

$$F_{di} = 3280 \text{ J}^{1/2} \text{ cm}^{3/2} \text{ mol}^{-1}$$

$$F_{pi} = 1 \times 490 = 490 \text{ J}^{1/2} \text{ cm}^{3/2} \text{ mol}^{-1} \text{ from } -\text{COO}-$$

$$E_{hi} = 1 \times 7000 = 7000 \text{ J/mol from } -\text{COO}-$$

$$\delta = \left( \sigma_a^2 + \sigma_p^2 + \sigma_h^2 \right)^{1/2} = \left[ (16.83)^2 + (2.51)^2 + (5.99)^2 \right]^{1/2}$$

$$\delta = 18.04 \text{ MPa}^{1/2}$$