

UNITED STATES PATENT AND TRADEMARK OFFICE

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BEFORE THE PATENT TRIAL AND APPEAL BOARD

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BAKER HUGHES, INC.,  
Petitioner

v.

LUBRIZOL SPECIALTY PRODUCTS, INC.,  
Patent Owner

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Case IPR2016- 00734  
Patent No. 8,022,118

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

Baker Hughes Incorporated

**Exhibit 1005**

BAKER HUGHES v. LUBRIZOL

IPR2016-00734

LSPI Ex. 2026  
IPR2016-01896

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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 Inc. (“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 petition for *inter partes* review concerns U.S. Patent No. 8,022,118 B2 (“the 118 patent”) and requests that the United States Patent and Trademark Office (USPTO) cancel claims 1-11 of the 118 patent.

3. Claims 1-11 of the 118 patent are directed generally to a method of reducing the frictional pressure loss associated with the turbulent flow of a heavy crude oil through a pipeline or other conduit by introducing a drag reducing polymer into the crude oil. The claims specify certain properties of both the heavy crude oil and the drag reducing polymer, which I will discuss below.

4. Throughout this declaration, I will refer to drag reducing polymers as the relevant field or the relevant art.

5. In preparing this Declaration, I have reviewed the 118 patent and considered each of the documents cited in Section III in light of the general

knowledge in the relevant art in 2005 more than one year before the earliest filing date of the 118 patent, i.e., before December 22, 2005. For ease of reference, every reference herein to 2005 means the time period in 2005 that is more than one year before the earliest filing date of the 118 patent. 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 March 1, 2016) *curriculum vitae* is provided as Exhibit 1006. 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 and Biomolecular Engineering with a joint appointment

in the Department of Materials Science and Engineering from 2011 to 2012. In 2012, I was promoted to Associate Professor with tenure at UD in the Department of Chemical and Biomolecular Engineering and Department of Materials Science and Engineering, which is my current position. I was awarded the chaired position of the Thomas and Kipp Gutshall Associate Professor of Chemical and Biomolecular Engineering in 2012, and I retain that appointment currently. My other academic appointments are listed in my *curriculum vitae*. Exhibit 1006.

9. One of the courses that I teach as an Associate 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 address is drag reducing additives and polymer flow properties in solution. Additionally, the course addresses polymer synthesis and polymer solubility as assessed through 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 and group contribution theory origins and methods.

10. Accordingly, I am an expert in the field of polymers and thermodynamics, and I have specific knowledge and understanding in the field of polymer flow properties in solution, including the field of polymeric drag reducing additives.

11. 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 the field at that time and was familiar with the knowledge and education of others working in the field.

### III. Documents Considered in Forming My Opinions

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

Exhibit	Description
1001	U.S. Patent No. 8,022,118 (“the 118 patent”)
1002	U.S. Patent No. 6,015,779 (“Eaton”)
1003	Strausz, O.P. <i>et al.</i> , <i>The Chemistry of Alberta Oil Sands, Bitumens and Heavy Oils</i> , Alberta Energy Res. Inst., Calgary, 2003 (“Strausz”)
1004	U.S. Patent No. 4,983,186 (“Naiman”)
1007	<i>Polymer Handbook</i> , 4 <sup>th</sup> ed., Brandrup, J. <i>et al.</i> (Eds.), John Wiley & Sons, Inc., New York, 1999 (“Brandrup”)
1008	Zakin, J.L. <i>et al.</i> (1980) “Effect of Polymer Molecular Variables on Drag Reduction” <i>J. Macromol. Sci.-Phys.</i> B18(4):795-814
1009	Mowla, D. <i>et al.</i> (2006) “Experimental Study of Drag Reduction by a Polymeric Additive in Slug Two-Phase Flow of Crude Oil and Air in Horizontal Pipes” <i>Chem. Eng. Sci.</i> 61:1549-54
1010	Material Safety Data Sheet for KOCH SURE SOL®-150 (2000)
1011	Barrufet, M. <i>et al.</i> (2003) “Reliable Heavy Oil-Solvent Viscosity Mixing Rules for Viscosities Up to 450 K, Oil-Solvent Viscosity Ratios Up to $4 \times 10^5$ , and Any Solvent Proportion” <i>Fluid Phase Equilibria</i> 213:65-79
1012	Fetters, J. <i>et al.</i> (1999) “Chain Dimensions and Entanglement Spacings in Dense Macromolecular Systems” <i>J. Polym. Sci. B: Polym. Phys.</i> 37:1023-33

Exhibit	Description
1033	Fetters, L. J. <i>et al.</i> “Chain Dimensions and Entanglement Spacings” in Mark, J.E. (Ed.), <i>Physical Properties of Polymers Handbook</i> , 2 <sup>nd</sup> ed., pp. 447-54, Springer Science+Business Media, Woodbury, NY, 2007
1034	Tait, P.J.T. <i>et al.</i> (1970) “Thermodynamic Studies on Poly( $\alpha$ -olefin)-Solvent Systems” <i>Polymer</i> 11:359-73
1035	<i>Polymers: A Property Database</i> , 2 <sup>nd</sup> ed., Ellis, B. <i>et al.</i> (Eds.), CRC Press, Boca Raton, FL, 2009
1036	<i>Handbook of Thermoplastics</i> , 2 <sup>nd</sup> ed., Olabisi, O. <i>et al.</i> (Eds.), CRC Press, Boca Raton, FL, 2016
1037	Sharp, D.G. <i>et al.</i> (1950) “Size and Density of Polystyrene Particles Measured by Ultracentrifugation” <i>J. Biol. Chem.</i> 185:247-53

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

13. 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, familiar with the conventional wisdom in the art as of that date, and a person of ordinary creativity.

14. 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, the common types of polymerization processes, types of polymerization catalysts, and solubility properties of polymers. He or she would regularly have consulted standard reference materials in the field, such as Brandrup *et al.*, *Polymer Handbook*, 4th ed., John Wiley & Sons, Inc., 1999 (Ex. 1007; “Brandrup”).

15. 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 Strausz, O.P. and Lown, E.M., *The Chemistry of Alberta Oil Sands, Bitumens and Heavy Oils*, Alberta Energy Research Institute, Calgary, 2003. (Ex. 1003; “Strausz”), 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. 1008; “Zakin”), and 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. 1009; “Mowla”).

16. Thus, such a person would have been readily able to determine properties such as the solubility parameters of polymers, including by such well-known techniques as group contribution theory calculations, enabling him or her to identify polymers that would be suitable for use as drag reducing additives in a target fluid.

17. I have been asked to analyze the 118 patent 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 application for the 118 patent was filed.



18. In my opinion, in 2005 a person of ordinary skill in the art relating to the subject matter of the 118 patent 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 or polymer flow properties in solution for any fluid.

19. 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. The 118 Patent**

20. The 118 patent, entitled “Drag Reduction of Asphaltenic Crude Oils” concerns reducing the pressure drop associated with the formation of turbulent eddies in crude oil having a high asphaltene content and a low API gravity flowing through a pipeline or other conduit. Ex. 1001. This reduction in pressure drop is achieved by treating the crude oil with a drag reducing polymer, more particularly a drag reducing polymer having a high molecular weight and a solubility parameter within about 20% of the solubility parameter of the crude oil. Ex. 1001 at Abstract.

21. As described by the 118 patent, fluids in a pipeline must be transported with a sufficient fluid pressure to achieve the desired throughput. Drops in fluid pressure, however, are often caused by turbulent eddies, such as those that can be initiated by 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.

22. As described by the 118 patent, 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 suppress the growth of turbulent eddies, and this suppression results in a higher flow rate of hydrocarbon liquid at a constant pumping pressure. Ex. 1001 at 1:28-39.

23. The 118 patent also states that 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. For example, as stated by the 118 patent, effective drag reducing agents “typically have molecular weights in excess of five million.” Ex. 1001 at 1:41-43.

24. According to the 118 patent, 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.

25. As a result, the 118 patent describes “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 1:44-49. The claims of the 118 patent reflect the goal of meeting this stated need. Specifically, each of the claims recites the step of introducing a drag reducing polymer into a liquid hydrocarbon having an API gravity of less than about 26° and an asphaltene content of at least 3 weight percent. Ex. 1001 at claims.

26. Asphaltenes are defined by the 118 patent 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 exists [sic] in crude oil.” Ex. 1001 at 2:64-3:8.

27. API gravity is defined by the 118 patent 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. 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.

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

TABLE 1

Asphaltene Content, API Gravity, and Elasticity Response								
Crude Oil		ASPHALTENE		ELASTICITY RESPONSE				
				(AFFINITY)				
		CONTENT		API	LP	LP		
Sample	Type	Test 1	Test 2	Gravity	100	300	Polymer A	Polymer B
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

29. The drag reducing polymer is defined in the claims of the 118 patent in terms of (a) its molecular weight, (b) its number of repeat units (which is related to molecular weight), and (c) its solubility parameter. Ex. 1001 at claims.

30. According to the 118 patent, 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 claim 4, claim 10. Yet, the 118 patent states that drag reducing polymers at that time typically had molecular weights in excess of five million g/mol. Ex. 1001 at 1:41-43.

31. According to the 118 patent, the drag reducing polymer has a solubility parameter that is similar to the solubility parameter of heavy crude oil. See Ex. 1001 at claim 1 (“the drag reducing polymer has a solubility parameter within  $4 \text{ MPa}^{1/2}$  of the solubility parameter of the liquid hydrocarbon”); Abstract (describing the drag reducing polymer as having “a solubility parameter within about 20% of the solubility parameter of the heavy crude oil”); compare 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 12:24-31 (describing the drag reducing polymer as having a solubility parameter within identical ranges).

32. A person of ordinary skill in the art would have known in 2005 that the purpose of solubility parameters is to predict or estimate the solubility of one compound in another, or more generally, the potential of two compounds to mix, as opposed to separate into two phases. For example, the closer the solubility parameters of two materials, the more likely to form a homogenous solution of those two materials. Accordingly, by stating that a drag reducing polymer and a certain crude oil have similar solubility parameters, the 118 patent would have simply indicated to a person of ordinary skill in the art in 2005 that the drag

reducing polymer was predicted (or estimated) to be soluble in the crude oil. Ex. 1001 at 1:39-41.

33. A person of ordinary skill in the art in 2005 would have known and understood the methods for calculating solubility parameters set forth in the 118 patent.

34. The 118 patent discloses that the solubility parameter of the liquid hydrocarbon is determined according to 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. Indeed, both this method for calculating solubility parameters and the Strausz publication referenced by the 118 patent would have been known by a person of ordinary skill in the art in 2005. See Ex. 1003 at 465-468; see also Ex. 1007 at 676.

35. Similarly, the 118 patent discloses that the solubility parameter of the drag reducing polymer is determined according to the Van Krevelen method of the Hansen solubility parameters 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. Both the equations in columns 11 and 12 of the 118 patent and the *Polymer Handbook* referenced by the 118 patent would have been known to a person of ordinary skill in the art in 2005. See Ex. 1007 at 677-683; see also Ex. 1003 at

469. A person of ordinary skill in the art in 2005 also would have recognized that the 118 patent contains a typographical error in the equation for calculating  $\delta_h$  in claims 7 and 11, in which the hydrogen-bonding term incorrectly states “ $F_{hi}$ ” instead of the correct “ $E_{hi}$ ”. The “F” terminology refers to a force and is appropriate for the polar and dispersive terms, while the “E” terminology refers to bond energy.

## **VI. Drag Reducing Polymers**

36. The problem of friction-related drops in fluid pressure when transporting any fluid, for example crude oil, was well known to a person of ordinary skill in the art in 2005. 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, naphthalenic, and alkane-based molecules. The non-linear nature of the aromatics and naphthalenics does not enable those molecules to dampen the turbulent eddies, leading to significant pressure drops. These pressure drops correspond to a reduction in fluid flowrate through the pipeline.

37. As described by the 118 patent, 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.

38. Those of ordinary skill in the art in 2005 would have known that drag reducing polymers were understood to lessen eddy-related drops in fluid pressure through a physical process. Drag reducing polymers 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.

39. 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 a few 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.

40. 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 composition and 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 properties of the crude oil, and those of ordinary skill in the art in 2005 would have known that result to be commercially undesirable.

41. 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. 1008 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. 1009 at 1550.

42. 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.

## **VII. The Eaton Patent**

43. United States Patent No. 6,015,779, entitled “methods for forming amorphous ultra-high molecular weight polyalphaolefin drag reducing agents,” identifying Gerald B. Eaton as the first-named inventor, issued on January 18, 2000. Ex. 1002 (“Eaton”). Eaton describes the preparation of a polyalphaolefin drag reducing agent and the use of the polyalphaolefin drag reducing agent in highly viscous crude oil. Because of its subject matter, a person of ordinary skill in the art in 2005 would have been familiar with the Eaton patent.

44. Eaton discloses the addition of a polymeric drag reducing agent to a hydrocarbon flowing through a pipeline, particularly viscous crude oil flowing through a pipeline, to improve the hydrocarbon flow. Ex. 1002 at 1:66-2:9.

45. According to Eaton, the polymeric drag reducing agents “interact with the turbulent flow processes and reduce frictional pressure losses such that the pressure drop for a given flow rate is less . . . .” Ex. 1002 at 1:29-33. More particularly, Eaton explains that the polymeric drag reducing agents “reduce the impact of turbulence through direct interaction and absorption of some or most [frictionally generated] energy bursts thus improving flow characteristics in the conduit.” Ex. 1002 at 5:8-23. Energy bursts of the sort described by Eaton are understood to cause turbulent eddies. Accordingly, in 2005 a person of ordinary skill in the art would have understood that the addition of the polymeric drag

reducing agents of Eaton would serve to suppress the growth of turbulent eddies, and thus reduce the frictional pressure loss associated with the turbulent flow of crude oil through a pipeline.

46. Eaton describes the polymeric drag reducing agents as overcoming the problems with commercially available polymers, which “are deficient when used with *highly viscous* crude oil, where the need may be the greatest.” Ex. 1002 at 1:48-52 (emphasis added). Eaton describes the improved polymeric drag reducing agents as being particularly effective for incorporation into *viscous* crude oil flowing through pipelines. Ex. 1002 at 1:66-2:9 (emphasis added). Highly viscous crude oil would generally have been understood by a person of ordinary skill in the art in 2005 to include heavy crude oil – e.g., the type of crude oil that the inventors of the 118 patent allege to have treated for the first time.

47. In Example 1, Eaton provides a description and data showing that his polymeric drag reducing agents are in fact effective when introduced into heavy crude oil of the sort described in the 118 patent. Specifically, Eaton describes a comparative test between two polyalphaolefin drag reducing agents made in accordance with his teaching (Compositions “A” and “B”) and two commercially available drag reducing compositions (Compositions “C” and “D”). Ex. 1002 at 13:55-14:5. Each of the four compositions was introduced into test hydrocarbons, and the percent flow increase was measured. Ex. 1002 at 14:33-36.

48. One of the test hydrocarbons is identified as Bow River crude oil, which is described by Eaton as being a “highly viscous” crude oil. Ex. 1002 at 14:36-38. In fact, as demonstrated by Table 1 of the 118 patent, reproduced above in paragraph 25, Bow River crude oil was known to a person of ordinary skill in the art, in 2005, to have an asphaltene content between 10.3 and 11.4 weight percent and an API Gravity of about 21.8°. Thus, Bow River crude oil meets the limitations of the liquid hydrocarbon recited in the claims of the 118 patent (an asphaltene content of at least 3 weight percent and an API Gravity of less than about 26°).

49. Compositions A and B, i.e., the two drag reducing agents prepared by Eaton, were found to reduce the frictional pressure loss associated with the turbulent flow of the Bow River crude oil, resulting in percent flow increases of 3.0% and 5.5%, respectively, when added at a concentration of 4.6 ppmw. See Table 1 of Eaton, reproduced below.

TABLE I			
<u>PERCENT FLOW INCREASE</u>			
DRA	ANS @3 ppm	BOW RIVER @4.6 ppm	HEXANE @1 ppm
A	15.0	3.0	40.1
B	12.0	5.5	37.5
C <sup>1</sup>	10.8	-0.5	31.1
D <sup>2</sup>	—	0.0	27.1

<sup>1</sup>LIQUID POWER™ commercial DRA from Conoco Inc.

<sup>2</sup>FLO-1005™ commercial DRA from Baker-Hughes, Inc.

50. A person of ordinary skill in the art would have understood in 2005 that the introduction of the drag reducing polymers in Compositions A and B to Bow River crude oil at the stated concentration (4.6 ppmw) as described in Example 1 would not appreciably reduce the viscosity of the heavy crude oil. The reason is that a person of ordinary skill in the art in 2005 would have known that the drag reducing polymers themselves, which are “ultra-high molecular weight” (Ex. 1002 at 14:6-31), would have no appreciable effect on the viscosity of the heavy crude oil when added at concentrations of parts per million to the crude oil.

51. Moreover, while each of Compositions A and B includes a carrier solvent, a person of ordinary skill in the art in 2005 would have recognized that the amount of solvent introduced into the Bow River crude oil from Compositions A and B would be small enough to have no appreciable effect on the viscosity of the crude oil. In particular, in 2005 a person of ordinary skill in the art would have known that the amount of solvent that is introduced into the Bow River crude oil from Compositions A and B can be calculated in the manner explained in the following paragraphs.

52. Composition A is described as being made from a reaction mixture containing 64,800 ppmw C<sub>6</sub> and ppmw 64,800 C<sub>12</sub> alpha-olefin monomers and 857,991 ppmw KOCH Sure-Sol-150 solvent. Ex. 1002 at 14:7-19. The monomers are polymerized to make the drag reduction polymer, which remains dissolved in

the solvent. Table 1 indicates that DRA (Drag Reducing Agent) A was added to Bow River crude oil at a concentration of about 4.6 ppmw of the polymer. A person of ordinary skill in the art in 2005 would have known that the concentration of solvent added to the Bow River crude oil with the polymer can thus be determined using the simple ratio:

$$\frac{129,600 \text{ ppmw DRA in Comp A}}{4.6 \text{ ppmw DRA in oil}} = \frac{857,991 \text{ ppmw solvent in Comp A}}{X \text{ ppmw solvent in oil}}$$

$$X \approx 30.5 \text{ ppmw}$$

53. Composition B is described as being made from a reaction mixture containing 39,500 ppmw C<sub>8</sub> and 39,500 ppmw C<sub>14</sub> alpha olefin monomers and 907,950 ppmw KOCH Sure-Sol-150 solvent. Ex. 1002 at 14:20-31. Table 1 indicates that DRA (Drag Reducing Agent) B was added to Bow River crude oil at a concentration of about 4.6 ppmw of the polymer. A person of ordinary skill in the art in 2005 would have known that the concentration of solvent added to the Bow River crude oil with the polymer can thus be determined using the simple ratio:

$$\frac{79,000 \text{ ppmw DRA in Comp B}}{4.6 \text{ ppmw DRA in oil}} = \frac{907,950 \text{ ppmw solvent in Comp B}}{X \text{ ppmw solvent in oil}}$$

$$X \approx 52.9 \text{ ppmw}$$

54. The solvent that is present in Compositions A and B is identified as KOCH Sure-Sol-150 solvent, which is mostly “Heavy Aromatic Solvent Naphtha.” Ex. 1010. At least in part because naphtha is also a component of crude oil, a person of ordinary skill in the art would have understood that the addition of up to 53 ppmw of KOCH Sure-Sol-150 solvent would not have an appreciable effect on the viscosity of the Bow River crude oil. A person of ordinary skill in the art could have confirmed his or her understanding by applying mixing rules, such as the Arrhenius mixing rule, which is applicable at extremely low concentrations of solvent, such as 53 ppm or less. (It is worth noting that more complicated mixing rules, such as those presented in Barrufet et al., *Reliable heavy oil-solvent viscosity mixing rules for viscosities up to 450 K, oil-solvent viscosity ratios up to  $4 \times 10^5$ , and any solvent proportion*, Fluid Phase Equilibria 213, pages 65-79, published in 2003 (Ex. 1011) (“Barrufet”), reduce to the Arrhenius mixing rule at the low solvent concentrations discussed herein). For instance, by applying the Arrhenius mixing rule to a KOCH Sure-Sol-150 solvent/heavy crude oil mixture and performing a calculation such as that presented as Attachment A, Calculation 1, a person of ordinary skill in the art would have recognized that the addition of 53 ppmw of KOCH Sure-Sol-150 to heavy crude oil has no appreciable effect on the viscosity of the crude oil.

55. In 2005, a person of ordinary skill in the art would have understood that the addition of between 30 ppmw and 53 ppmw of KOCH Sure-Sol-150 solvent with a drag reducing polymer to Bow River heavy crude oil would not appreciably decrease the viscosity of the crude oil. Accordingly, in 2005, a person of ordinary skill in the art would have understood that the introduction of Compositions A and B to Bow River crude oil, as described in Example 1 of Eaton, would not measurably reduce the viscosity of the heavy crude oil.

56. Eaton also describes the addition of a drag reducing polymer without a carrier solvent. Ex. 1002 at 11:43-67. If a solvent is not used during the polymerization process, Eaton explains, a polyalphaolefin block is formed. Ex. 1002 at 11:43-56. The polyalphaolefin block then may be used to reduce drag in a conduit by adding the polyalphaolefin block or pieces of the polyalphaolefin block to the conduit. Ex. 1002 at 11:57-67. Accordingly, if in 2005 a person of ordinary skill in the art had been concerned about the introduction of solvent impacting the composition of the crude oil, such as by reducing the viscosity of the crude oil, that person would simply have followed Eaton's teaching to introduce the drag reducing polymer without a carrier solvent (which would not reduce the viscosity of the crude oil).

57. According to Eaton, flow can be improved when a drag reducing polymer is added to a liquid hydrocarbon at a concentration as low as 1 part per



million by weight. Ex. 1002 at 2:6-9. More particularly, Eaton teaches that the drag reducing polymers should be added to a pipeline having viscous crude oil at a concentration of from about 1 to about 250 ppmw by weight, and more preferably from about 25 to 150 ppmw by weight. Ex. 1002 at 2: 32-39. The amounts disclosed by Eaton are well within the about 0.1 to about 500 ppmw by weight range claimed by the 118 patent.

58. Eaton also describes certain properties of the drag reducing polymers prepared according to his disclosure. For instance, Eaton states that the drag reducing polymer must have an “ultra-high molecular weight,” which is defined as a molecular weight corresponding to an inherent viscosity of at least about 10 dL/g. Ex. 1002 at 6:15-19. According to Eaton, this corresponds roughly to a molecular weight of at least 10 or 15 million g/mol. Ex. 1002 at 6:29-32. Preferably, the ultra-high molecular weight polymers of Eaton have molecular weights of greater than 25 million g/mol. Ex. 1002 at 6:32-34. Eaton also states that the polymers should have a narrow molecular weight distribution. Ex. 1002 at 6:34-36. Accordingly, the drag reducing polymers disclosed by Eaton have weight average molecular weights that are well above the 1 million ( $1 \times 10^6$ ) g/mol lower boundary claimed by the 118 patent.

59. Eaton does not explicitly disclose the numbers of repeat units in the drag reducing polymers (note that while the 118 patent recites the term “repeating

units”, I prefer to use the technically accurate term “repeat units” and have used it throughout this Declaration in place of “repeating units”). In 2005, however, a person of ordinary skill in the art easily could have determined that the drag reducing polymers of Eaton contain in excess of 25,000 repeat units.

60. Specifically, the drag reducing polymers of Eaton are described as comprising alpha olefin monomers such as “ethylene, propylene, 1-butene, 4-methyl-1-pentene, 1-hexene, 1-octene, 1-decene, 1-dodecene and 1-tetradecene; conjugated or unconjugated dienes such as butadiene and 1,4-hexadiene; aromatic vinyls such as styrene; and cyclic olefins such as cyclobutene.” Ex. 1002 at 9:6-25. Additionally, the drag reducing polymers are described as having a molecular weight of at least 10 million g/mol.

61. For a polyalphaolefin to have a molecular weight of 10 million g/mol and 25,000 repeat units, the units that make up the polymer would have an average molecular weight per repeat unit of 400 g/mol. Each of the alpha olefin monomers listed by Eaton has a molecular weight significantly less than 400 g/mol. Accordingly, the preferred polymers disclosed by Eaton would necessarily have more than 25,000 repeat units to achieve a molecular weight of 10 million g/mol.

62. Using the molecular weights of the repeat units present in the polymer of Composition A ( $C_6$  and  $C_{12}$ ), which are taught by Eaton as being present in a 1:1 weight ratio, and assuming a molecular weight of the polymer of 10 million g/mol

(the lower bound disclosed by Eaton), I have calculated that the polymer of Composition A has greater than 89,000 repeat units. My calculation is presented below:

$$\frac{5 \text{ million g/mol polymer}}{84.16 \text{ g/mol C}_6} = 59,411 \text{ repeat C}_6 \text{ units}$$

$$\frac{5 \text{ million g/mol polymer}}{168.32 \text{ g/mol C}_{12}} = 29,705 \text{ repeat C}_{12} \text{ units}$$

$$59,411 \text{ repeat C}_6 \text{ units} + 29,705 \text{ repeat C}_{12} \text{ units} = 89,116 \text{ repeat units}$$

63. Similarly, using the molecular weights of the repeat units present in the polymer of Composition B (C<sub>8</sub> and C<sub>14</sub>), which are taught by Eaton as being present in a 1:1 weight ratio, and assuming a molecular weight of the polymer of 10 million g/mol (the lower bound disclosed by Eaton), I have calculated that the polymer of Composition B has greater than 70,000 repeat units. My calculation is presented below:

$$\frac{5 \text{ million g/mol polymer}}{112.22 \text{ g/mol C}_8} = 44,555 \text{ repeat C}_8 \text{ units}$$

$$\frac{5 \text{ million g/mol polymer}}{196.38 \text{ g/mol C}_{14}} = 25,461 \text{ repeat C}_{14} \text{ units}$$

$$44,555 \text{ repeat C}_8 \text{ units} + 25,461 \text{ repeat C}_{14} \text{ units} = 70,016 \text{ repeat units}$$

64. Though Eaton makes no explicit mention of the solubility parameter of any drag reducing polymer, the Eaton disclosure provided ample information from which a person of ordinary skill in the art could have calculated the solubility parameters of the disclosed drag reducing polymers.

65. A person of ordinary skill in the art in 2005 would have been able to identify the solubility parameters of the various drag reducing polymers disclosed by Eaton. For instance, the solubility parameters of many common polymers were published in well-known handbooks, such as the *Polymer Handbook* referenced by the 118 patent (i.e., Brandrup). Ex. 1007 at VII-702 to VII-711. Additionally, it was well understood in 2005 that one could calculate the solubility parameter of a polymer using published solubility parameters for the incorporated monomer units (the “repeat units”), such as from Brandrup, and group contribution equations. Ex. 1007 at VII-686.

66. For instance, I have calculated the solubility parameter of Composition A of Eaton using the group contribution equations presented in Brandrup at B11 and C30, i.e., the equations recited at col. 11, line 47 to col. 12, line 23 of the 118 patent, as shown on page 1 of Attachment B, and arrived at a solubility parameter value of  $16.31 \text{ MPa}^{1/2}$ . See also Attachment C for the values and calculations used in determining this solubility parameter; Ex. 1033 at 449

(identifying the density of 1-hexene, “a-PHEX”); Ex. 1034 at 363 (identifying the density of polydodecene-1).

67. I also have calculated the solubility parameter of Composition B of Eaton using the group contribution equations presented in Brandrup at B11 and C30, i.e., the equations recited at col. 11, line 47 to col. 12, line 23 of the 118 patent, as shown on page 2 of Attachment B, and arrived at a solubility parameter value of 16.11 MPa<sup>1/2</sup>. See also Attachment C for the values and calculations used in determining this solubility parameter; Ex. 1012 at 1025 (identifying the density of octene, “a-POCT”); Ex. 1035 at 895 (identifying the density of tetradecene).

68. A person of ordinary skill in the art would recognize that (because the only interactions are dispersive interactions) the solubility parameter for a liquid hydrocarbon, such as Bow River heavy crude oil, would follow the equation  $\delta_2 = [(\Delta H_v - RT)/V)]^{1/2}$ . A person also would understand, however, that the solubility parameter for a crude oil could not be directly calculated using this equation because not all of the components of the crude oil would vaporize (and thus components would lack a  $\Delta H_v$ ). Accordingly, a person of ordinary skill in the art attempting to determine the solubility parameter of a crude oil using this equation would be led to look to alternative methods such as solvent fractionation experiments. Specifically, because asphaltene is one of the highest molecular weight fractions of the crude oil (and thus plays a principal role in determining the

boundaries of oil solubility characteristics), a person of ordinary skill in the art attempting to use this equation to determine the solubility parameter of heavy crude oil would be led to solvent fractionation experiments involving asphaltene. A summary of several experiments is provided by *The Chemistry of Alberta Oil Sands, Bitumens and Heavy Oils*, written by Otto P. Strausz and Elizabeth M. Lown, and published by Alberta Energy Research Institute in 2003. Ex. 1003 at 467-468.

69. The results summarized by Strausz demonstrate that asphaltene is completely soluble in solvents having solubility parameters as low as about 17.1 MPa<sup>1/2</sup> and as high as about 22.1 MPa<sup>1/2</sup>. Ex. 1003 at 467-468. According to Strausz, in other words, to completely solubilize asphaltene, the solubility parameter of a material must be within about 2.5 MPa<sup>1/2</sup> of the solubility parameter of the asphaltene (which is identified by Strausz as 19.6 MPa<sup>1/2</sup> for purposes of calculating this range). Because asphaltene is fully solubilized by Bow River crude oil, a person of ordinary skill in the art would have recognized from the solvent fractionation experiments summarized in Strausz, which use the equation  $\delta_2 = [(\Delta H_v - RT)/V]^{1/2}$ , that the solubility parameter of Bow River crude oil must fall within a range between 17.1 MPa<sup>1/2</sup> and 22.1 MPa<sup>1/2</sup>.

70. For the reasons explained below, a person of ordinary skill in the art also would have recognized from the solvent fractionation experiments

summarized by Strausz that Compositions A and B of Eaton have solubility parameters that fall within about  $3.5 \text{ MPa}^{1/2}$  of the solubility parameter of the Bow River crude oil.

71. From the experimental results provided at Table 14.1 of Strausz, a person of ordinary skill in the art would recognize that for asphaltene to be reasonably soluble in a solvent, the solvent must have a solubility parameter that is within about  $3.5 \text{ MPa}^{1/2}$  of the solubility parameter of the asphaltene. Ex. 1003 at 468. Specifically, Table 14.1 demonstrates that asphaltene is reasonably soluble in solvents having a solubility parameter of  $16.2 \text{ MPa}^{1/2}$  (with only about 1 wt. % of the bitumen reported as precipitate) whereas asphaltene is not reasonably soluble in solvents having a solubility parameter of  $16.0 \text{ MPa}^{1/2}$  (with 8.5 wt. % of the bitumen reported as precipitate). Therefore, a person of ordinary skill in the art would recognize that the lower boundary solubility parameter at which asphaltene is reasonably soluble in a material is at about  $16.1 \text{ MPa}^{1/2}$ . In other words, for asphaltene to be reasonably soluble in a material, that material must have a solubility parameter that is within about  $3.5 \text{ MPa}^{1/2}$  of the solubility parameter of asphaltene (which is identified by Strausz as about  $19.6 \text{ MPa}^{1/2}$ ).

72. Both the Athabasca bitumen and the materials used as solvents in the solvent fractionation experiments summarized by Strausz are hydrocarbon-based. Accordingly, because the solubility parameters of the hydrocarbons can be

estimated using only the dispersive interactions, a person of ordinary skill in the art would have understood that the numerical relationships demonstrated by Strausz also would apply to other hydrocarbons, as the physics of these interactions are translatable across systems [as indicated by Strausz, p. 466]. For instance, because the Bow River crude oil of Eaton is primarily hydrocarbon-based, a person of ordinary skill in the art would have understood that to be reasonably soluble in Bow River crude oil, a hydrocarbon material must have a solubility parameter that is within about  $3.5 \text{ MPa}^{1/2}$  of the solubility parameter of the crude oil.

73. The polyalphaolefin polymers of Eaton Compositions A and B are also hydrocarbons. Moreover, because the polyalphaolefin polymers of Eaton Compositions A and B were reported to achieve drag reduction in Bow River crude oil, a person of ordinary skill in the art would have recognized that the polyalphaolefin polymers of Compositions A and B are at least reasonably soluble in the Bow River crude oil. Indeed, because solubility is necessary for drag reduction, if Compositions A and B of Eaton were not at least reasonably soluble in the Bow river crude oil, no drag reduction would have been reported.

74. Because a person of ordinary skill in the art would have recognized (a) that the numerical relationship between solubility parameters understood from Strausz would apply to Compositions A and B and the Bow River crude oil of Eaton, and (b) that Compositions A and B are demonstrated by Eaton as being



reasonably soluble in Bow River crude oil, a person of ordinary skill in the art would have understood that Eaton Compositions A and B have solubility parameters that are within about  $3.5 \text{ MPa}^{1/2}$  of the solubility parameter of the Bow River crude oil.

75. A person of ordinary skill in the art would therefore have understood that, using the calculation methods disclosed in Brandrup and Strausz, the solubility parameter for the drag reducing polymers of Compositions A and B of Eaton are within the range of  $4 \text{ MPa}^{1/2}$  of the solubility parameter of the Bow River crude oil into which they are introduced, as recited in claim 1 of the 118 patent.

76. I have been asked to compare claims 1, 3-4, 6-7, and 11 of the 118 patent to the disclosure of Eaton. It is my opinion that a person of ordinary skill in the art would have understood in 2005 that Eaton discloses each and every element of claims 1, 3-4, 6-7, and 11 of the 118 patent.

### **VIII. The Strausz Book**

77. 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. 1003 (“Strausz”). Strausz addressed a number of topics related to the composition and properties of heavy crude oils. Ex. 1003 at 464-480.

78. Because of its subject matter and its publication prior to 2005, a person of ordinary skill in the art considering drag reduction for liquid hydrocarbons would have been familiar with Strausz. Indeed, Strausz is cited by the 118 patent itself for its description of solubility parameters for liquid hydrocarbons. Ex. 1001 at 4:17-36.

79. Strausz generally describes the use of solubility parameters in the context of heavy crude oil. For example, Strausz explains that “[t]he ability of a solvent to solubilize asphaltene or, in general, to dissolve a solid or to form a homogenous solution with another liquid, may be expressed in terms of solubility parameters.” Ex. 1003 at 465. After introducing an equation for calculating a Hildebrand solubility parameter, designated  $\delta_2$ , Strausz states that “liquids differing by two Hildebrands in their  $\delta$  values are generally incompletely miscible.” Ex. 1003 at 466.

80. Strausz also states that “the correlation between the solubility of asphaltene and solvent solubility parameter is quite good for nonpolar and low-polarity solvents.” Ex. 1003 at 466. Indeed, Strausz describes an exhaustive study of the correlation between asphaltene solubility and solubility parameters, which demonstrates 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. 1003 at 467. In other words, to be an effective

solvent for asphaltene, Strausz teaches that a material should have a solubility parameter between  $17.1 \text{ MPa}^{1/2}$  and  $22.1 \text{ MPa}^{1/2}$ , *i.e.*, a solubility parameter within about  $2.5 \text{ MPa}^{1/2}$  of the solubility parameter of asphaltene (which is identified as being about  $19.6 \text{ MPa}^{1/2}$ ).

## **IX. The Combination of Eaton and Strausz**

81. As described above, in 2005, a person of ordinary skill in the art would have recognized that the more soluble a drag reducing polymer becomes in the fluid being treated, the more effective that drag reducing polymer would be at reducing turbulent frictional losses. In 2005, a person of ordinary skill in the art also would have understood that the solubility of one material in another might routinely be predicted by comparing the measured or calculated solubility parameters of the two compounds. Specifically, for example, it was well understood in 2005 that the closer the solubility parameters of two materials, the more likely one would obtain a homogenous (one-phase) solution of the two materials. See, *e.g.*, Ex. 1003 at 465-467.

82. Moreover, to maximize the effectiveness of the drag reducing polymer of Eaton, a person of ordinary skill in the art would have been motivated to prepare the drag reducing polymer of Eaton to have a solubility parameter that is as close as possible to the solubility parameter of the crude oil into which it is intended for use. For instance, in 2005, a person of ordinary skill in the art would have been

motivated to prepare the drag reducing polymer of Eaton so as to have a solubility parameter that is as close as possible to the solubility parameter of highly viscous Bow River crude oil.

83. On the basis of the combination of Eaton and Strausz, a person of ordinary skill in the art in 2005 would have been able to determine that the solubility parameter of the Bow River crude oil lies between about  $17.1 \text{ MPa}^{1/2}$  and about  $19.6 \text{ MPa}^{1/2}$ . As described above (see paragraphs 66-67), the solubility parameter for Composition B of Eaton can be calculated to have a value of  $16.1 \text{ MPa}^{1/2}$ . Because the solubility parameter of Composition B would be understood as being within about  $3.5 \text{ MPa}^{1/2}$  of the solubility parameter of Bow River crude oil (because Composition B is demonstrated as being reasonably soluble in the crude oil, see paragraphs and 70-74), the solubility parameter of the Bow River crude oil would be understood as being about  $19.6 \text{ MPa}^{1/2}$  or less. Moreover, because the Bow River crude oil would be understood as fully solubilizing asphaltene (which makes up at least 10 wt. % of the crude oil), the solubility parameter of Bow River crude oil would be understood as being about  $17.1 \text{ MPa}^{1/2}$  or greater (see paragraph 69). Therefore, by simply applying these two conditions, a person of ordinary skill in the art would have understood that the solubility parameter of the Bow River crude oil disclosed by Eaton should be between about  $17.1 \text{ MPa}^{1/2}$  and about  $19.6 \text{ MPa}^{1/2}$ .

84. Accordingly, it is my opinion that, in 2005, a person of ordinary skill in the art would have been motivated by Eaton's disclosure of successfully incorporating drag reducing polymers in heavy, high-asphaltene Bow River crude oil to prepare and utilize a drag reducing polymer having a solubility parameter within the range of about 17.1 to about 19.6 MPa<sup>1/2</sup>, as determined using the teachings of Eaton and Strausz, because it would be particularly effective in heavy, high-asphaltene crude oil such as Bow River crude oil.

85. Using the disclosure of Eaton, in 2005, a person of ordinary skill in the art would have been able to prepare a drag reducing polymer having a solubility parameter between 17.1 MPa<sup>1/2</sup> and 19.6 MPa<sup>1/2</sup>. For instance, in 2005, a person of ordinary skill would have needed only to (a) identify, such as by calculating from published solubility parameter information such as that within Brandrup, a desirable homopolymer or copolymer having a solubility parameter within the target range and then (b) perform the polymerization reaction under conditions that are suitable to obtain the desired homopolymer or copolymer.

86. One of the monomers disclosed by Eaton as being used as a repeat unit in the drag reducing polymers is styrene. Ex. 1002 at 9:8-18. A person of ordinary skill in the art could easily have identified the solubility parameter of polystyrene as being at least about 17.45 MPa<sup>1/2</sup> by simply finding it within the

published solubility parameter information in Table 10 of Brandrup. Ex. 1007 at VII-707.

87. Alternatively, a person of ordinary skill in the art could have identified the solubility parameter of polystyrene by performing a conventional group contribution calculation, such as using the group contribution equations presented in Brandrup at B11 and C30, i.e., the equations recited at col. 11, line 47 to col. 12, line 23 of the 118 patent. For instance, using Brandrup, which was available in 2005, I calculated the solubility parameter of polystyrene as follows:

- First, the molar volume of each repeat unit is calculated by dividing the molecular weight of an incorporated monomer segment (or “repeat unit”) by its density at a specific temperature (the density is most often reported at 25 °C). For example, the molecular weight of the styrene repeat unit is 104.15 g/mol [which could easily have been obtained in 2005 through a routine calculation or from the available literature], and its density at 25 °C is 1.05 g/cm<sup>3</sup> [which could readily be found in 2005 in conventional resources such as polymer handbooks and the available peer-reviewed scientific literature, see for example Ex. 1037 at 252-253 (identifying the density of polystyrene)], which gives a molar volume of about 99.19 cm<sup>3</sup>/mol.

- Second, the types of functional groups in the repeat unit are determined by reference to Table 4 of Brandrup. Ex. 1007 at VII-686. In the case of polystyrene, this corresponds to 1-CH<sub>2</sub>, 1-CH, and 1-phenyl ring.
- Third, the dispersive (F<sub>di</sub>), polar (F<sub>pi</sub>), and hydrogen-bonding (E<sub>hi</sub>) contributions of each functional group are determined from Table 4 of Brandrup. In the case of polystyrene, these values are  
 1-CH<sub>2</sub>: F<sub>di</sub> = 270, F<sub>pi</sub> = 0, and E<sub>hi</sub> = 0;  
 1-CH: F<sub>di</sub> = 80, F<sub>pi</sub> = 0, and E<sub>hi</sub> = 0, and  
 1-phenyl ring: F<sub>di</sub> = 1430, F<sub>pi</sub> = 110, and E<sub>hi</sub> = 0.  
 Note that the units for F<sub>di</sub> and F<sub>pi</sub> are J<sup>1/2</sup>cm<sup>3/2</sup>mol<sup>-1</sup>, and the units for E<sub>hi</sub> are J/mol.
- Fourth, the above values are entered into the equations listed at C30 in Brandrup to calculate the solubility parameter component contributions. Ex. 1007 at VII-683. In the case of polystyrene, these values are  $\delta_d = 17.95$  MPa<sup>1/2</sup>,  $\delta_p = 1.11$  MPa<sup>1/2</sup>, and  $\delta_h = 0.00$  MPa<sup>1/2</sup>.
- Fifth, the solubility parameter component values are entered into the equation listed at B11 in Brandrup to calculate the solubility parameter. Ex. 1007 at VII-677. In the case of polystyrene, this gives a solubility parameter of  $\delta = 17.98$  MPa<sup>1/2</sup>. See Attachment C for the values and calculations used in determining this solubility parameter.

88. Accordingly, based on the target solubility parameter range between  $17.1 \text{ MPa}^{1/2}$  and  $19.6 \text{ MPa}^{1/2}$ , a person of ordinary skill in the art in 2005 seeking to provide a drag reducing polymer for Bow River crude oil would have been motivated to prepare, and could easily have prepared using the process of Eaton, a polymer for use as a drag reducing additive by preparing a polystyrene polymer, which would have a solubility parameter of about  $17.98 \text{ MPa}^{1/2}$ .

89. Alternatively, based on the target solubility parameter range between  $17.1 \text{ MPa}^{1/2}$  and  $19.6 \text{ MPa}^{1/2}$ , a person of ordinary skill in the art in 2005 seeking to provide a drag reducing polymer for Bow River crude oil would have been motivated to and easily could have prepared a polymer for use as a drag reducing additive by generating a poly(ethylene/styrene) copolymer through a process such as Ziegler Natta polymerization of ethylene [Ex. 1002 at 9:17] and styrene [Ex. 1002 at 9:18]. If the monomer feed were selected to include at least 45 wt.% styrene, the resulting polymer would have an estimated solubility parameter of at least  $17.1 \text{ MPa}^{1/2}$ , as calculated using the group contribution methods described in Brandrup.

90. For instance, using information that was available in 2005, *e.g.*, Brandrup, I calculated the solubility parameter of polystyrene as set forth above in paragraph 87. Using the same process, I calculated the solubility parameter of polyethylene as set forth below:



- First, the molar volume of the monomeric repeat unit is calculated by dividing the molecular weight of an incorporated monomer (or “repeat unit”) by its density at a specific temperature (the density is most often reported at 25 °C). For example, the molecular weight of the ethylene repeat unit is 28.05 g/mol [which could easily have been obtained in 2005 through a routine calculation or from the available literature], and its density at 25 °C is 0.851 g/cm<sup>3</sup> [which could readily be found in 2005 in conventional resources such as polymer handbooks and the available peer-reviewed scientific literature, see for example Ex. 1012 at 1025 (identifying the density of ethylene, “PE”)], which gives a molar volume of about 32.96 cm<sup>3</sup>/mol.
- Second, the types of functional groups in the repeat unit are determined by reference to Table 4 of Brandrup. Ex. 1007 at VII-686. In the case of polyethylene, this corresponds to 2-CH<sub>2</sub>.
- Third, the dispersive (F<sub>di</sub>), polar (F<sub>pi</sub>), and hydrogen-bonding (E<sub>hi</sub>) contributions of each functional group are determined from Table 4 of Brandrup. In the case of polyethylene, the values for 2-CH<sub>2</sub> are F<sub>di</sub> = 540 (270 x 2), F<sub>pi</sub> = 0, and E<sub>hi</sub> = 0. Note that the units for F<sub>di</sub> and F<sub>pi</sub> are J<sup>1/2</sup>cm<sup>3/2</sup>mol<sup>-1</sup>, and the units for E<sub>hi</sub> are J/mol.

- Fourth, the above values are entered into the equations listed at C30 in Brandrup to calculate the solubility parameter component contributions. Ex. 1007 at VII-683. In the case of polyethylene, these values are  $\delta_d = 16.38 \text{ MPa}^{1/2}$ ,  $\delta_p = 0 \text{ MPa}^{1/2}$ , and  $\delta_h = 0 \text{ MPa}^{1/2}$ .
- Fifth, the solubility parameter component values are entered into the equation listed at B11 in Brandrup to calculate the solubility parameter. Ex. 1007 at VII-677. In the case of polyethylene, this gives a solubility parameter of  $\delta = 16.38 \text{ MPa}^{1/2}$ . See Attachment C for the values and calculations used in determining this solubility parameter.

91. Because the solubility parameter of the polystyrene repeat units is  $17.98 \text{ MPa}^{1/2}$  and the solubility parameter of the polyethylene repeat units is  $16.38 \text{ MPa}^{1/2}$ , a person of ordinary skill in the art would have understood that by varying the relative amounts of each monomer in the reaction mixture, one could prepare a poly(ethylene/styrene) copolymer having a particular solubility parameter from within the range of  $16.38 \text{ MPa}^{1/2}$  and  $17.98 \text{ MPa}^{1/2}$ . For example, in order to prepare a poly(ethylene/styrene) copolymer having a solubility parameter of  $17.1 \text{ MPa}^{1/2}$ , a person of ordinary skill in the art could easily have determined that the copolymer should contain about 45 wt.% polystyrene units using the equation:

$$w_{PE}\delta_{PE} + w_{PS}\delta_{PS} = 17.1 \text{ MPa}^{1/2}$$

in which  $w_{PE}$  is the weight percent of polyethylene units,  $w_{PS}$  is the weight percent of polystyrene units (noting that the sum of  $w_{PE}$  and  $w_{PS}$  must equal 1),  $\delta_{PE}$  is the solubility parameter of polyethylene, and  $\delta_{PS}$  is the solubility parameter of polystyrene.

92. In 2005, the process described in paragraphs 85-91 would have been a matter of routine for a person of ordinary skill in the art. Indeed, by 2005, a person of ordinary skill in the art would generally have known how to perform this process to obtain polymers having desired solubilities in particular solvents.

93. Through a simple comparison of solubility parameters, a person of ordinary skill in the art would have understood that polymers having solubility parameters between  $17.1 \text{ MPa}^{1/2}$  and  $17.98 \text{ MPa}^{1/2}$ , such as those described above, would be within  $4 \text{ MPa}^{1/2}$ , and more particularly within  $2.5 \text{ MPa}^{1/2}$ , of the solubility parameter of the Bow River crude oil into which the drag reducing polymers were introduced, as is recited in the claims of the 118 patent.

94. It is my opinion that in 2005, a person of ordinary skill in the art would have had a reasonable expectation that, by combining the teachings of Eaton and Strausz, one could have successfully prepared a drag reducing polymer having particularly desirable properties, including the claimed solubility parameter relationship, for reducing frictional pressure losses in Bow River heavy crude oil. As explained above, in 2005, a person of ordinary skill in the art seeking to provide

a drag reducing polymer for heavy crude oil would have had both a motivation to consult the teachings of Eaton and Strausz, and a reasonable expectation that, by combining these teachings and using one's ordinary skill, one would have prepared a drag reducing polymer that was effective for reducing frictional pressure losses in Bow River crude oil.

95. I have been asked to compare claims 1-7 and 11 of the 118 patent to the combination of Eaton and Strausz. It is my opinion that a person of ordinary skill in the art would have understood in 2005 that the combination of Eaton and Strausz discloses each and every element of claims 1-7 and 11 of the 118 patent.

#### **X. The Naiman Patent**

96. United States 4,983,186, entitled "Methods and Compositions for Reduction of Drag in Hydrocarbon Fluids," identifying Michael I. Naiman as the first-named inventor, issued on January 8, 1991. Ex. 1004 ("Naiman"). Naiman describes a method for reducing friction loss in flowing hydrocarbon fluids by adding a polymer of styrene, an alkyl acrylate, and a carboxylic acid. Ex. 1004 at Abstract. Because of its subject matter, in 2005, a person of ordinary skill in the art would have been familiar with the Naiman patent.

97. Naiman describes his invention as relating to pressure loss as a result of friction in the flow of hydrocarbons, and particularly hydrocarbon fuels, through conduits, and to additives for the reduction of such frictional pressure loss. Ex.

1004 at 1:7-12. Specifically, for example, Naiman describes the problems associated with frictional pressure loss during the long distance transport of crude oil extracted from Alaskan reserves through pipelines. Ex. 1004 at 1:14-27.

98. To reduce this frictional pressure loss, Naiman describes a drag reducing agent that has both sufficient molecular weight to be effective and solubility in the hydrocarbon fluid. Ex. 1004 at 2:6-14. In fact, because “the effectiveness of the polymer increases with its oil-solubility,” preferred polymers are described as being oil-soluble at concentrations that are significantly higher than those encountered in drag-reducing applications. Ex. 1004 at 3:56-62. The preferred polymers also are described as tending to be elongate, rather than “balling up.” Ex. 1004 at 3:56-62. Additionally, the polymer of Naiman is said to have a molecular weight of about 3 million g/mol to about 5 million g/mol (prior to any polyamine salt formation). Ex. 1004 at 3:47-49.

99. Naiman describes a drag-reducing polymer that is prepared from styrene, an alkyl acrylate, and, in most cases, a carboxylic acid using an emulsion polymerization process. The styrene component is said to increase the molecular weight of the polymer, while the alkyl acrylate is said to provide the polymer with improved oil solubility. Ex. 1004 at 2:56-62. It was well known in 2005 to a person of ordinary skill in the art that alkyl acrylates contain oxygen atoms.

100. Naiman does not explicitly disclose the numbers of repeat units in the drag reducing polymers. In 2005, however, a person of ordinary skill in the art easily could have determined that many of the drag reducing polymers of Naiman contain in excess of 25,000 repeat units.

101. Specifically, the drag reducing polymers of Naiman are described as comprising styrene, alkyl acrylates of from about four to about twelve carbon atoms (and preferably from about six to about ten carbon atoms), and, in some cases, a small amount of acrylic or methacrylic acid. Ex. 1004 at 2:56-3:17. Additionally, the drag reducing polymers are described as having a molecular weight of at about 3,000,000 to about 5,000,000 g/mol. A person of ordinary skill in the art would recognize, based on the identity of the units that make up the polymer and the molecular weight of the polymer, that many of the polymers disclosed by Naiman have greater than 25,000 repeating units.

102. For example, using polymer 5 from Example 1 of Naiman as a basis, I easily was able to calculate that the polymer would have greater than 25,000 repeat units so long as the molecular weight of the polymer was above about 3.84 million (Naiman teaches that the polymers have a molecular weight between 3-5 million).

The calculation is provided below:

MW styrene = 104.15 g/mol; MW 2-ethylhexyl acrylate = 184.28 g/mol  
wt. % styrene = 38.13 %; wt. % 2-ethylhexyl acrylate = 61.87 %

X g/mol polymer

$$\frac{\text{-----}}{(104.15 \text{ g/mol} \times 0.3813) + (184.28 \text{ g/mol} \times 0.6187)} = 25,000 \text{ repeat units}$$

$$X = 3,843,101$$

103. If, for instance, the 2-ethylhexyl acrylate were replaced with butyl acrylate (which has seven carbon atoms as opposed to eleven), a polymer having the same weight percentages of Example 1, polymer 5, would have greater than 25,000 repeat units across the entire range of molecular weights (3-5 million) disclosed by Naiman. The calculation is provided below:

MW styrene = 104.15 g/mol; MW butyl acrylate = 128.17 g/mol  
wt. % styrene = 38.13 %; wt. % butyl acrylate = 61.87 %

3,000,000 g/mol polymer

$$\frac{\text{-----}}{(104.15 \text{ g/mol} \times 0.3813) + (128.17 \text{ g/mol} \times 0.6187)} = 25,208 \text{ repeat units}$$

In summary, a person of ordinary skill in the art would have recognized that there are many ways in which drag reducing polymers having greater than 25,000 units could be prepared following the disclosure of Naiman.

104. According to Naiman, the polymer also may be combined with a polyamine to form an oil-soluble salt of higher effective molecular weight to produce an especially effective high molecular weight and oil soluble drag reducing agent. Specifically, Naiman discloses that a carboxylic acid may be introduced into the polymer and then the acidic polymer and a basic polyamine

may be reacted to form a salt. By doing so, Naiman reports an ability to increase the molecular weight of the drag reducing polymer beyond that achieved through the polymerization alone (between 3 g/mol and 5 million g/mol) without causing a significant decrease in solubility. Ex. 1004 at 4:31-37; see also 3:8-17 (noting that because the carboxylic acid tends to decrease the solubility of the polymer, it is incorporated as a very small percentage of the polymer, such as less than 1% by weight).

105. According to Naiman, superior results were achieved when the drag reducing polymer is added to hydrocarbon fuel, such as crude oil, at a concentration from about 3 ppmw to about 35 ppmw. Ex. 1004 at 4:12-21.

106. In Example 2, Naiman explains that the polymers of his disclosure are effective at reducing frictional pressure losses in hydrocarbon liquids, whether used alone or in combination with a polyamine. For instance, Naiman provides the results of an experiment in which various (a) polymers and (b) combinations of polymers (acidic polymer and polyamine) were introduced into kerosene, and the percentage of drag reduction (% D.R.) achieved over the untreated fluid was measured. Ex. 1004 at 5:21-55. Tables I and II demonstrate that, when added to kerosene at a concentration of 100 ppmw, polymers consisting of about 62-63 wt.% 2-ethyl-hexyl acrylate (EHA) and about 37-38 wt.% styrene (St.) achieved a drag reduction of about 21-26%. Ex. 1004 at Tables I and II (see polymers 1 and



5). When similar polymers were rendered acidic and reacted with a polyamine, they were able to achieve a drag reduction between 19% and about 39%. Ex. 1004 at Table II.

## **XI. The Combination of Eaton, Strausz, and Naiman**

107. As described above, based on the teachings of Eaton and Strausz, in 2005, a person of ordinary skill in the art would have recognized that, to be most effective in the Bow River crude oil disclosed by Eaton, a drag reducing polymer should have a solubility parameter between  $17.1 \text{ MPa}^{1/2}$  and  $19.6 \text{ MPa}^{1/2}$ .

108. Although a person of ordinary skill in the art would have understood that it was possible to prepare an effective drag reducing polymer using the disclosure of Eaton, the polymerization process disclosed by Eaton – *i.e.*, conventional Ziegler-Natta polymerization – was limited in terms of monomer choices and required the use of an expensive catalyst system. A person of ordinary skill in the art, in 2005, would therefore have been led to look to other known drag reducing polymers that would be effective in heavy crude oil such as the Bow River crude oil disclosed by Eaton, and that might be prepared using a more accessible and/or more economical polymerization process. A person of ordinary skill in the art would thus have been led to Naiman.

109. Naiman describes a drag reducing polymer that is effective when used in crude oil. The polymers disclosed by Naiman are made up of (1) styrene and

alkyl acrylate units or (2) styrene, alkyl acrylate, and carboxylic acid units that then are reacted with a polyamine (although the carboxylic acid and the polyamine are present in the polymer at such low amounts that they will have little effect on the solubility parameter of the polymer when complexed into a macromolecular salt). By simply identifying the solubility parameters of the repeat styrene and alkyl acrylate units, such as by using a solubility parameter table of the sort provided in Brandrup and performing a group contribution calculation, a person of ordinary skill in the art would have recognized that the polymers of Naiman would typically have solubility parameters that are within the target range of  $17.1 \text{ MPa}^{1/2}$  to  $19.6 \text{ MPa}^{1/2}$ . Accordingly, a person of ordinary skill in the art would have recognized that the drag reducing polymers of Naiman would be soluble, and thus effective, in the Bow River heavy crude oil disclosed by Eaton.

110. Moreover, the polymers of Naiman are described as being prepared by a conventional emulsion polymerization process. A person of ordinary skill in the art would have understood that the emulsion polymerization process of Naiman would be (a) less limited in terms of monomers that may be used and (b) employ a less expensive catalyst system than the Ziegler-Natta polymerization process described by Eaton. Accordingly, in 2005, a person of ordinary skill in the art would have considered the drag reducing polymers of Naiman to be an

advantageous replacement for the polyalphaolefin-derived materials of Eaton for use in Bow River heavy crude oil.

111. Therefore, it is my opinion that in 2005 a person of ordinary skill in the art seeking to provide a drag reducing polymer for incorporation into Bow River crude oil would have been motivated to replace the drag reducing polymers of Eaton with one of the drag reducing polymers of Naiman.

112. Using the disclosure of Naiman, in 2005, a person of ordinary skill in the art would readily have been able to prepare a drag reducing polymer having a solubility parameter that is within the target range of  $17.1 \text{ MPa}^{1/2}$  to  $19.6 \text{ MPa}^{1/2}$ . For instance, in 2005, a person of ordinary skill would need only to (a) identify, such as by performing a conventional group contribution calculation, which of the copolymers of Naiman have a solubility parameter within the target range, and (b) perform the polymerization reaction under conditions that are suitable to obtain the desired copolymer.

113. Using the group contribution equation of Brandrup, which was available in 2005, I easily was able to calculate the solubility parameters for a number of polymers of Naiman. I calculated the solubility parameter of polystyrene units as described above in paragraph 87. By employing the same framework, I also calculated the solubility parameters of a number of the alkyl acrylate repeat units described in Naiman. For instance, I calculated the solubility

parameter of poly(hexyl acrylate) to be  $\delta = 17.36 \text{ MPa}^{1/2}$ , the solubility parameter of poly(octyl acrylate) to be  $\delta = 17.52 \text{ MPa}^{1/2}$ , and the solubility parameter of poly(dodecyl acrylate) to be  $\delta = 17.95 \text{ MPa}^{1/2}$ . See Attachment C for the values and calculations used in determining these solubility parameters; Ex. 1036 at 180 (identifying the densities of hexyl acrylate and dodecyl acrylate, i.e. “Poly(lauryl acrylate”)); Ex. 1033 at 450 (identifying the density of octyl acrylate, “a-POA”). The other alkyl acrylates described by Naiman would be expected to have similar solubility parameters (the above samples demonstrating that the solubility parameters of the alkyl acrylates increase slightly as the number of carbon atoms increases).

114. Accordingly, a polymer that was made up of styrene and hexyl acrylate repeat units would have a solubility parameter between about  $17.36 \text{ MPa}^{1/2}$  and  $17.98 \text{ MPa}^{1/2}$ , depending on the relative amounts of each monomer that was incorporated. Similarly, a polymer that was made up of styrene and octyl acrylate repeat units would have a solubility parameter between about  $17.52 \text{ MPa}^{1/2}$  and  $17.98 \text{ MPa}^{1/2}$ , depending on the relative amounts of each monomer that was incorporated. Similarly, a polymer that was made up of styrene and dodecyl acrylate repeat units would have a solubility parameter between about  $17.95 \text{ MPa}^{1/2}$  and  $17.98 \text{ MPa}^{1/2}$ , depending on the relative amounts of each monomer that was incorporated. Note also that a person of ordinary skill in the art would have

understood that the inclusion of a carboxylic acid and a polyamine, as is taught in some embodiments of Naiman, would have had little impact on the solubility parameter of the drag reducing polymer, according to the group contribution methods of Brandrup, because of the small amounts in which they are incorporated (the carboxylic acid being less than about 1 wt. %).

115. Because most, if not all, of the polymers disclosed by Naiman have a solubility parameter within the desired range, a person of ordinary skill in the art would have been able to prepare a drag reducing polymer that was desirable for use in Bow River crude oil simply by following the emulsion polymerization process described by Naiman, for instance using any of the combinations of monomers described above. By a simple comparison of solubility parameters, a person of ordinary skill in the art would have understood that the solubility parameter of the resulting drag reducing polymers would be within  $4 \text{ MPa}^{1/2}$ , and more particularly within  $2.5 \text{ MPa}^{1/2}$ , of the solubility parameter of the Bow River crude oil, as is recited in the claims of the 118 patent.

116. It also would have been obvious to a person of ordinary skill in the art to have prepared the drag reducing polymer of Naiman to have greater than 25,000 repeat units, so as to provide a drag reducing polymer having a relatively high molecular weight, which is described by Naiman, and generally understood by persons of ordinary skill in the art, to be desirable for a drag reducing polymer.

117. Accordingly, it is my opinion that in 2005 a person of ordinary skill in the art would have had a reasonable expectation that, by combining the teachings of Eaton, Strausz, and Naiman, one would have successfully prepared a drag reducing polymer in which a plurality of the repeat units comprise an oxygen heteroatom and which has the properties recited in the claims of the 118 patent, including a solubility parameter that is within  $4 \text{ MPa}^{1/2}$  of the solubility parameter of the Bow River crude oil disclosed by Eaton.

118. As I have explained above, in 2005, a person of ordinary skill in the art would have had both a motivation to consult the teachings of Eaton, Strausz, and Naiman, and a reasonable expectation that, by combining these teachings, one could have prepared a drag reducing polymer that was particularly effective for reducing frictional pressure losses in Bow River heavy crude oil.

119. For these same reasons, a person of ordinary skill in the art would have been motivated to incorporate the drag reducing polymers taught by Naiman into the Bow River heavy crude oil disclosed by Eaton.

120. In doing so, a person of ordinary skill in the art would have recognized that the drag reducing polymer of Naiman should be added to the Bow River heavy crude oil in an amount between about 3 and about 35 ppmw by weight, which is the range that is described by Naiman as providing superior drag reduction results. Ex. 1004 at 4:12-16. This range also coincides with the range

described by Eaton, which teaches the addition of drag reducing polymer to highly viscous crude oil in amounts between 1 and 250 ppmw, and preferably between 25 and 150 ppmw. Ex. 1002 at 2:35-40.

121. By incorporating the drag reducing polymers of Naiman into the Bow River heavy crude oil disclosed by Eaton, a person of ordinary skill in the art would have understood that the drag reducing polymer of Naiman would cause a reduction in frictional pressure loss associated with turbulent flow by suppressing the growth of turbulent eddies. This is, at least in part, because the drag reducing polymers of Naiman would be understood to have properties, *e.g.* (a) a straight chain structure with minimal branching, (b) high molecular weight, and (c) solubility in the Bow River crude oil, which would make it an effective drag reducing agent in Bow River crude oil.

122. A person of ordinary skill in the art would also have recognized how to incorporate the drag reducing polymer of Naiman into Bow River crude oil, such as in the amounts described above, in a manner that would not measurably decrease the viscosity of the crude oil.

123. For example, in Example 2, Naiman describes drying the drag reducing polymers and then dissolving them in xylene for addition to the subject test fluid (in that example, kerosene). Ex. 1004 at 5:21-25. As long as the amount of xylene solvent was kept at a reasonable level, *e.g.*, about 105 ppmw xylene per

35 ppmw of polymer (which corresponds with the dilution levels for the emulsion polymerization suggested by Naiman at 4:16-21), a person of ordinary skill in the art would have understood that addition of the solution would not measurably decrease the viscosity of Bow River crude oil. For example, by applying the mixing rules of Barrufet (Ex. 1011) and performing a calculation such as that presented in Attachment A, Calculation 2, a person of ordinary skill in the art would have recognized that those amounts of xylene would have no appreciable effect on the viscosity of a heavy crude oil.

124. Alternatively, to the extent that one might be concerned with the introduction of solvent affecting the properties of the Bow River crude oil, it would have been obvious to a person of ordinary skill in the art to have incorporated the drag reducing polymer as a solid, such as is taught by Eaton (e.g., by adding the dried polymer disclosed in Example 2 of Naiman directly to the crude oil). Ex. 1002 at 11:43-67. A person of ordinary skill in the art in 2005 would have known that the drag reducing polymers themselves would have no appreciable effect on the viscosity of the heavy crude oil when added at concentrations of parts per million to the crude oil.

125. I have been asked to compare claims 8-10 of the 118 patent to the combination of Eaton, Strausz and Naiman. In my opinion, a person of ordinary skill in the art would have understood in 2005 that the combination of Eaton,



Strausz, and Naiman discloses each and every element of claims 8-10 of the 118 patent.

## **XII. Conclusion**

126. 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.

Respectfully submitted,



Date: 3/31/2016

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Thomas H. Epps, III, Ph.D.

# ATTACHMENT A

### Calculation 1

using Arrhenius mixing rule  $\rightarrow$  standard for low conc. systems

$$\log \mu = x_o \log \mu_o + x_s \log \mu_s$$

$o = \text{oil}$  ;  $s = \text{solvent}$

$$\bar{m}_o = 710.32 \text{ g/mol} \rightarrow \text{see Barratet, page 67}$$

$$\rho_o = 0.946 \text{ g/cm}^3 \quad \text{Table 1 for "Oil A" as estimate}$$

$$\bar{m}_s = 128.17 \text{ g/mol} \rightarrow \text{assuming naphthalene; lower bound}$$

$$\rho_o = 0.9 \text{ g/cm}^3 (15.6^\circ\text{C}) \quad \text{MSDS for KOCH Sure-Sol-150}$$

$$53 \text{ ppmw} = 0.0053 \text{ weight \% or } 5.3 \times 10^{-5} \text{ weight fraction}$$

$$x_s = \frac{\text{mol}_s}{\text{mol}_{\text{total}}} = \frac{5.3 \times 10^{-5} \text{ g} / 128.17 \text{ g mol}^{-1}}{1 \text{ g} / 710.32 \text{ g mol}^{-1}} = 2.937 \times 10^{-4}$$

using 1g basis

$$\log \mu = 1 \times \log(2152 \text{ cP}) + 2.937 \times 10^{-4} \times \log(0.964 \text{ cP})$$

$$\log \mu = 3.33284 \Rightarrow \underline{\mu = 2152 \text{ cP}}$$

using  $\mu_o = 2152 \text{ cP}$  at  $295.7 \text{ K} (23^\circ\text{C}) \rightarrow$  see Barratet  
 $\mu_s = 0.964 \text{ cP}$  at  $80^\circ\text{C}$  p.73, Table 5

$$\underline{\mu_o = \mu = 2152 \text{ cP}} \quad \checkmark$$

## Calculation 2

page 1

105 ppmw = 0.0105 weight % or  $1.05 \times 10^{-4}$  weight fraction

$$\log \mu = \left( \frac{\alpha V_o}{\alpha V_o + V_s} \right) \log \mu_o + \left( 1 - \frac{\alpha V_o}{\alpha V_o + V_s} \right) \log \mu_s \quad \text{see ref.}$$

O = oil, S = solvent

$$V_o = \frac{m_o}{\rho_o(T)} \quad \text{and} \quad V_s = \frac{m_s}{\rho_s(T)}$$

$$\alpha = 0.3524 \times x_s^{-0.71154} \quad \text{see ref}$$

$$\bar{m}_o = 710.32 \text{ g/mol} \quad \text{Canada Alberta Heavy Crude}$$

$$\rho_o = 0.946 \text{ g/cm}^3 \quad \text{see ref}$$

$$\bar{m}_s = 106.16 \text{ g/mol}$$

$$\rho_s = 0.864 \text{ g/cm}^3$$

$$\bar{V}_o = \frac{710.32 \text{ g/mol}}{0.946 \text{ g/cm}^3} = 750.87 \text{ cm}^3/\text{mol}$$

$$\bar{V}_s = \frac{106.16 \text{ g/mol}}{0.864 \text{ g/cm}^3} = 122.87 \text{ cm}^3/\text{mol}$$

$$x_s = \frac{\text{mol}_s}{\text{mol}_{\text{total}}} = \frac{1.05 \times 10^{-4} \text{ g} / 106.16 \text{ g mol}^{-1}}{1 \text{ g} / 710.32 \text{ g mol}^{-1}} = 7.026 \times 10^{-4}$$

using 1g basis

## Calculation 2

Page 2

$$\alpha = 0,3524 \times X_5^{-0,71154} \quad \text{gives } \alpha > 1 \quad \text{for} \quad \text{see ref.}$$
$$X_5 = 7,026 \times 10^{-4}$$

$$\Rightarrow \alpha = 1$$

$$\log \mu = X_0 \log \mu_0 + X_5 \log \mu_5 \quad \text{reduces to}$$

Arrhenius mixing rule

$$\log \mu = 1 \times \log(2152 \text{ cP}) + 7,026 \times 10^{-4} \times \log(0,81 \text{ cP})$$

$$\log \mu = 3,3328 \Rightarrow \underline{\mu = 2152 \text{ cP}}$$

using  $\mu_0 = 2152 \text{ cP}$  at  $295,7 \text{ K}$  ( $23^\circ \text{C}$ )  $\rightarrow$  see ref.  
 $\mu_5 = 0,81 \text{ cP}$  at  $20^\circ \text{C}$  (o-xylene)

$$\underline{\mu_0 = \mu = 2152 \text{ cP}} \quad \checkmark$$



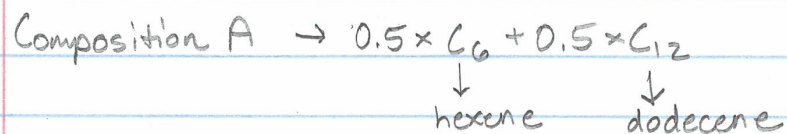
decane is basis for reference: assumption is that xylene can follow similar general relationship at extremely low mole fractions

ref  $\rightarrow$  Barrufet 2003

# ATTACHMENT B



page 1



polyhexene  $\rightarrow 84.16$  repeat unit MW [g/mol]  
PH  $\qquad \qquad 0.871$  is PH density at  $0^\circ\text{C}$  [g/cm<sup>3</sup>]

$$\bar{V}(\text{molar volume}) = \frac{84.16 \text{ g/mol}}{0.871 \text{ g/cm}^3} = 96.62 \text{ cm}^3/\text{mol}$$

PH units  $\rightarrow 1 \times \text{CH}_3 + 4 \times \text{CH}_2 + 1 \times \text{CH}$

$$F_{di} = 1 \times 420 + 4 \times 270 + 1 \times 80 = 1580 \text{ (J}^{1/2} \text{cm}^{3/2} \text{mol}^{-1})$$

$$F_{pi} = 0 ; E_{hi} = 0$$

$$\underline{\sigma_{PH} = 16.35 \text{ MPa}^{1/2}}$$

polydodecene  $\rightarrow 168.32$  repeat unit MW [g/mol]

PDD  $\qquad \qquad 0.856$  is PDD density at  $30^\circ\text{C}$  [g/cm<sup>3</sup>]

$$\bar{V} = \frac{168.32 \text{ g/mol}}{0.856 \text{ g/cm}^3} = 196.64 \text{ cm}^3/\text{mol}$$

PDD units  $\rightarrow 1 \times \text{CH}_3 + 10 \times \text{CH}_2 + 1 \times \text{CH}$

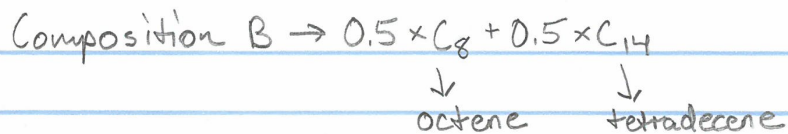
$$F_{di} = 1 \times 420 + 10 \times 270 + 1 \times 80 = 3200 \text{ (J}^{1/2} \text{cm}^{3/2} \text{mol}^{-1})$$

$$F_{pi} = 0 ; E_{hi} = 0$$

$$\underline{\sigma_{PDD} = 16.27 \text{ MPa}^{1/2}}$$

$$0.5 \times 16.35 \text{ MPa}^{1/2} + 0.5 \times 16.27 \text{ MPa}^{1/2} = \boxed{16.31 \text{ MPa}^{1/2}}$$

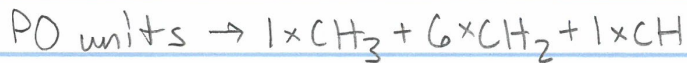
Page 2



polyoctene  $\rightarrow$  112.22 repeat unit MW [g/mol]

PO                      0.851 is PO density at 50°C [g/cm<sup>3</sup>]

$$\bar{V} = \frac{112.22 \text{ g/mol}}{0.851 \text{ g/cm}^3} = 131.87 \text{ cm}^3/\text{mol}$$



$$F_{di} = 1 \times 420 + 6 \times 270 + 1 \times 80 = 2120 \text{ (J}^{1/2} \text{cm}^{3/2} \text{mol}^{-1})$$

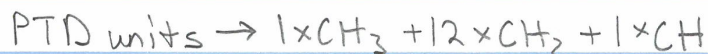
$$F_{pi} = 0 ; E_{hi} = 0$$

$$\underline{\delta_{PO} = 16.08 \text{ MPa}^{1/2}}$$

polytetradecene  $\rightarrow$  196.38 repeat unit MW [g/mol]

PTD                      0.848 is PTD density at 25°C [g/cm<sup>3</sup>]

$$\bar{V} = \frac{196.38 \text{ g/mol}}{0.848 \text{ g/cm}^3} = 231.58 \text{ cm}^3/\text{mol}$$



$$F_{di} = 1 \times 420 + 12 \times 270 + 1 \times 80 = 3740 \text{ (J}^{1/2} \text{cm}^{3/2} \text{mol}^{-1})$$

$$F_{pi} = 0 ; E_{hi} = 0$$

$$\underline{\delta_{PTD} = 16.15 \text{ MPa}^{1/2}}$$

$$0.5 \times 16.08 \text{ MPa}^{1/2} + 0.5 \times 16.15 \text{ MPa}^{1/2} = \boxed{16.11 \text{ MPa}^{1/2}}$$



# ATTACHMENT C

### Solubility Parameter Calculations

Monomer	Repeat unit <i>g/mol</i>	Density <i>g/cm<sup>3</sup></i>	Temperature <i>K</i>	Molar Volume <i>cm<sup>3</sup>/mol</i>	$\delta_d^{1/2}$ <i>MPa<sup>1/2</sup></i>	$\delta_p^{1/2}$ <i>MPa<sup>1/2</sup></i>	$\delta_h^{1/2}$ <i>MPa<sup>1/2</sup></i>	$\delta_{tot}^{1/2}$ <i>MPa<sup>1/2</sup></i>
ethylene	28.05	0.851	298	32.961	16.38	0.00	0.00	16.38
1-hexene	84.16	0.871	273	96.625	16.35	0.00	0.00	16.35
1-octene	112.22	0.851	323	131.868	16.08	0.00	0.00	16.08
1-dodecene	168.32	0.856	303	196.636	16.27	0.00	0.00	16.27
1-tetradecene	196.38	0.848	298	231.580	16.15	0.00	0.00	16.15
styrene	104.15	1.05	298	99.190	17.95	1.11	0.00	17.98
hexyl acrylate	156.22	0.98	298	159.408	15.75	3.07	6.63	17.36
octyl acrylate	184.3	0.98	298	188.061	16.22	2.61	6.10	17.52
dodecyl acrylate	240.38	0.99	298	242.808	17.01	2.02	5.37	17.95

## Density Values

Monomer	Temperature <i>K</i>	Density <i>g/cm<sup>3</sup></i>	Reference
ethylene	298	0.851	Fetters, L. J.; Lohse, D. J.; Graessley, W. W. J. Polym. Sci. Part B: Polym. Phys. 1999 37 (10): 1023-1033.
1-hexene	273	0.871	Fetters, L. J.; Lohse, D. J.; Colby, R. H. in Physical Properties of Polymers Handbook, 2nd ed; Mark, J. E., Ed.; AIP Press: Woodbury, NY, 2007; p 447-454.
1-octene	323	0.851	Fetters, L. J.; Lohse, D. J.; Graessley, W. W. J. Polym. Sci. Part B: Polym. Phys. 1999 37 (10): 1023-1033.
1-dodecene	303	0.856	Tait, P.J.T. and Livesey, P.J. <i>Polymer</i> 1970 11 (7): 359-373.
1-tetradecene	298	0.848	Polymers: A Property Database, Second Edition; Bryan Ellis, Ray Smith, Eds.; CRC Press: Boca Raton, FL, 2008.
styrene	298	1.05	Sharp, D.G. and Beard, J.W., Size and Density of Polystyrene Particles Measured by Ultracentrifugation, J. Biol. Chem. 1950, 185:247-253
hexyl acrylate	298	0.98	Handbook of Thermoplastics, Second Edition; Olagoke Olabisi, Kolapo Adewale, Eds.; CRC Press: Boca Raton, FL, 2016.
octyl acrylate	298	0.98	Fetters, L. J.; Lohse, D. J.; Colby, R. H. in Physical Properties of Polymers Handbook, 2nd ed; Mark, J. E., Ed.; AIP Press: Woodbury, NY, 2007; p 447-454.
dodecyl acrylate	298	0.99	Handbook of Thermoplastics, Second Edition; Olagoke Olabisi, Kolapo Adewale, Eds.; CRC Press: Boca Raton, FL, 2016.

## Group Contributions

Monomer	-CH3	-CH2-	-CH-	-C-	=CH2	=CH-	=C-	Benzyl	-COO-	Ring	-O-	-NH2	Fdi	Fpi	Fpi^2	Ehi
	A	B	C	D	E	F	G	H	I	J	K	L				
ethylene		2											540	0	0	0
1-hexene	1	4	1										1580	0	0	0
1-octene	1	6	1										2120	0	0	0
1-dodecene	1	10	1										3200	0	0	0
1-tetradecene	1	12	1										3740	0	0	0
styrene		1	1					1					1780	110	12100	0
hexyl acrylate	1	6	1						1				2510	490	240100	7000
octyl acrylate	1	8	1						1				3050	490	240100	7000
dodecyl acrylate	1	12	1						1				4130	490	240100	7000
$F_{di} (J^{1/2} \text{ cm}^{3/2} \text{ mol}^{-1})$	420	270	80	-70	400	200	70	1430	390	190	100	280				
$F_{pi} (J^{1/2} \text{ cm}^{3/2} \text{ mol}^{-1})$	0	0	0	0	0	0	0	110	490	0	400	0				
$E_{hi} (J \text{ mol}^{-1})$	0	0	0	0	0	0	0	0	7000	0	3000	84000				